ENGINEERING
LIBRARY

FEB 18 1948

NUCLEONICS

AN INTERNATIONAL JOURNAL
FOR TECHNIQUES AND APPLICATIONS OF NUCLEAR SCIENCE

February, 1948

ELECTRONICS FOR THE NUCLEAR PHYSICIST

OXYGEN-18 AS A TRACER ELEMENT

ISOTOPE TECHNIQUES IN BIOCHEMISTRY—V

THE NAVAL RESEARCH LABORATORY BETATRON

MANHATTAN DISTRICT DECLASSIFIED DOCUMENTS

RADIATION-MEASUREMENT BIBLIOGRAPHY

A McGraw-Hill Publication

NUCLEONICS

An International Journal for Techniques and Applications of Nuclear Science

Vol. 2, No. 2 · February, 1948

EDITORIAL BOARD

L. B. Borst
John R. Dunning
H. H. Goldsmith
Clark Goodman
F. C. Henriques, Jr.
W. F. Libby
L. D. Marinelli
I. Perlman
Edith H. Quimby
Winfield W. Salisbury
T. I. Taylor

Walter M. DeCew.....*Technical Editor*

Jerome D. Luntz.....*Assistant Editor*
Ethel Pelzer.....*Assistant Editor*

Robert Colborn.....*Washington Editor*
Gladys Montgomery.*Washington Editor*

Keith Henney.....*Consulting Editor*

E. J. Tangerman.....*General Manager*

NUCLEONICS (with which is consolidated ATOMIC POWER and ATOMIC ENGINEERING), January, 1948, Vol. 2, No. 2. Publication Office: 212 East York St., York, Pa. Published monthly by McGraw-Hill Publishing Company, Inc., 330 W. 42nd St. New York 18, N.Y. Application for entry as second-class matter at York, Pa., is pending, under the act of March 3, 1879. Copyright 1948 by McGraw-Hill Publishing Company, Inc. All rights reserved. James H. McGraw, Jr., President, J. A. Gerardi, Secretary, Curtis W. McGraw, Treasurer. Editorial and circulation offices, 330 W. 42nd St., New York 18, N.Y. Subscription rates, United States and Possessions, $15 for one year. Return postage guaranteed.

Contents

Electronics for the Nuclear Physicist..... 4
W. C. ELMORE

Oxygen-18 as a Tracer Element..... 18
RONALD BENTLEY

Manhattan District Documents..... 30

Isotope Techniques in Biochemistry—V..... 33
NORMAN S. RADIN

The Naval Research Laboratory Betatron..... 46
H. F. KAISER, D. L. MOCK, O. E. BERG, D. K. STEVENS and W. E. HARRIS

Bibliography: Radiation Measurement..... 63
MONICA HEALEA

Editorial..... 1
Nucleonic Events..... 66
Products and Materials..... 68
Abstracts..... 70

NUCLEONICS AN INTERNATIONAL JOURNAL
FOR TECHNIQUES AND APPLICATIONS OF NUCLEAR SCIENCE

Parallels between German Science before 1932 and Nucleonics in the United States Today

THE DEVELOPMENT OF THE SCIENTIFIC COMMUNITY at any given period of its history is generally reflected by the forms of publication it evolves to meet its needs. If we examine from this standpoint German scientific and industrial activities between 1913 and 1932, we are led to a number of revealing and instructive comparisons with the direction currently being taken by nuclear science and technology in the United States.

> In 1913 Germany was acknowledged to be the most scientifically advanced country in the world. The achievements of German science in the preceding few decades had been strongly reflected in the industrial development of the nation, and had brought great prestige to science and scientists.

The year 1913 also brought the founding of the then unique periodical *Die Naturwissenshaften*, which soon became an integral part of the scientific life of the period. *Die Naturwissenshaften* (the natural sciences) was published by the Julius Springer Company in Berlin, and edited until 1932 by Dr. Arnold Berliner, as a weekly international periodical for lengthy and detailed reviews of advances in science, medicine, and technology. One of the conditions in European science which brought about the founding of *Naturwissenshaften*, as quoted below from Berliner's editorial for the first issue, might equally well have been said for nuclear science and technology in 1948.

> "The rapidly increasing specialization in all domains of research," said Berliner in 1913, "makes it difficult for the individual investigator to orient himself beyond closely bordering fields. On the other hand, the more one is forced to narrow his own field of work, the more conscious he becomes of the intellectual need to comprehend the whole with its parts, and of his dependence on other fields of investigation."

With the object of counterbalancing the inevitable growth of specialization, *Naturwissenshaften* invited and made payment for review articles by the leading scientists of the day. In 1932, the year of Berliner's retirement, the

contributors included such investigators as Amaldi, Bethe, Debye, Einstein, Du Bridge, Nernst, Weyl, Placzek, Rossi, Heisenberg, and Rabinowitch.

> The structure and spirit of the flourishing scientific world reflected in *Naturwissenshaften* can now be seen starting to repeat itself in the United States, particularly if one looks closely at the social organization and historical orientation of Germany between 1913 and 1932. The main characteristic of scientific activity in this period was its emphasis in fundamental research, and the high social value attached to purely intellectual achievement. How this came about can be understood in terms of the generally held assumption that Germany had arrived earlier than the rest of industrial civilization at the stage in the development of its science when it is no longer economically sensible to distinguish between "pure" and "applied" research.

German industry sympathetically, patiently, and profitably supported a large fraction of fundamental research. Partly because industry paid large salaries and consulting fees, and partly because German laboratories were run quite undemocratically according to present American standards, outstanding workers were frequently able to retire as millionaires.

> The German Federal Government, through the Kaiser Wilhelm Association for the Advancement of Science, established and generously financed large government research centers in many cities—the world famous Kaiser Wilhelm Institutes. There were over fifteen such institutes, each organized for the investigation of a special field, as illustrated by the Kaiser Wilhelm-Institute für Chemie at Berlin-Dahlem, the Kaiser Wilhelm-Institute für Medizinisches Forschung at Heidelberg, and the Kaiser Wilhelm-Institute für Eisenforschung at Düsseldorf. Additional institutes were dedicated to such fields as physics, cell physiology, biochemistry, coal research, biophysics, and other special studies.

It takes little effort to see in the United States today a similar pattern of organization and outlook emerging in its development of nuclear science and technology. American private industrial enterprises, now conscious that "pure" science has dovetailed with the practical arts in the twentieth century, are investing in long-term basic nuclear research at such places as the Chicago Institute for Nuclear Studies, sponsoring fundamental chemical and physical investigations in company laboratories, and seeking wider opportunities to contribute to basic nuclear developments on the basis of enlightened self-interest. The National Bureau of Standards, and the Brookhaven, Clinton and Argonne National Laboratories give promise of achieving the national rôles and international eminence formerly held by the Kaiser Wilhelm Institutes in Germany.

Finally, since basic scientific advances can only be the product of disciplined and inspired minds, it is encouraging to note the gradual growth of the European and pre-1932 German public attitude toward science and scholarship which formed the environment that gave us investigators like Hilbert, Planck, Einstein, Warburg, Franck, Haber, Weyl, Pauli, and so many others of similar brilliance. These public attitudes took it for granted that all creative intellectual activities, artistic, literary, or scientific, were vital and integral parts of civilized life, and did not require their constant justification in terms of military, commercial or medical applications. In response to the slowly widening acceptance of similar attitudes in the United States, the new generation of scientists in government, university and industrial laboratories throughout the nation are turning to more basic, long-range investigations with increasing enthusiasm and self-confidence.

The progressive disintegration of German science that accompanied the rise of Nazism and loss of political freedom after 1932 was strikingly testified to by the declining quality of *Naturwissenshaften* after that date. Many of its leading contributors were leaving the country, others were losing their prerogatives for free research, and the articles that did appear grew ever more stale and insignificant. Few basic discoveries ever came out of the Third Reich, for the scientists who remained at work had surrendered their great traditions to the concept that social responsibility justifies restrictions on scientific freedom.

—*W. M. D.*

ELECTRONICS FOR THE NUCLEAR PHYSICIST

A thorough and authoritative presentation of theory and methods of design of fast electronic circuits. In this first article of a series, the use of the Laplace transformation in transient analysis of electrical networks is discussed

By W. C. ELMORE

Swarthmore College, Swarthmore, Pennsylvania

THE MAJORITY of measuring instruments used nowadays by the nuclear physicist involve electronic circuits. The intensive work on problems of instrumentation at various laboratories of the Manhattan Project, as well as related work on new or improved circuits for radar and other war-time applications (especially at the MIT Radiation Laboratory) has considerably refined and extended the state of the art. Indeed, if problems in electronic control, accelerators, cosmic-ray research, etc., are included, there exists today a body of knowledge on electronic instrumentation which would require at least a book-length exposition for its adequate presentation.

This series of review articles aspires to nothing so ambitious; in fact, it is nothing more than a revision of a set of notes written to supplement a series of sixteen lectures on "Fast Electronic Circuits" given by the author at Princeton University (during the spring term of 1947). The purpose of the lectures, now the purpose of the review, was to present a small part of basic electronic theory, and then to acquaint the listener with certain concepts underlying the design of a few specific circuits of great utility to the nuclear physicist. The lectures, in part, served as a commentary on electronic instruments designed at the Los Alamos Scientific Laboratory, and now employed extensively at various university laboratories.

On account of limitation in time and space, the number of types of circuits considered is far from complete. In particular no discussion is made of circuits used primarily with Geiger-Müller counters, nor is there any discussion of d-c amplifiers. The main emphasis is placed on the theory and the design of circuits for amplifying, recording and utilizing pulses from an ionization chamber or other proportional counter employing (fast) electron collection. Some of the most important advances in nuclear physics instrumentation during the war years have occurred in this general area, and it is hoped that the present exposition will prove helpful to the nuclear physicist who intends to apply war-born techniques to his current research. In addition it is hoped that the review will be of interest to the manufacturer of electronic devices, who may intend to design instruments for the nuclear physicist.

I. Method of the Laplace Transformation

Some knowledge of the transient behavior of electrical networks is necessary in designing and in understanding the operation of electronic circuits. The most convenient method of transient analysis is that based on the Laplace transformation. Although a complete treatment of this method would require considerable space for its presentation, it is possible in a brief,

introductory account to provide a means for handling the majority of simple transient problems likely to arise in electronic circuit design.

The typical problem which we shall consider is the following: a network consisting of inductances, capacitances, resistances and other elements whose operation is essentially linear (such as vacuum tubes operated class A) is quiescent prior to time $t = 0$. At this instant, a source of emf (or of current) becomes active in some part of the circuit. Currents then start to flow, voltages appear, etc., throughout the network. The source of excitation for the general case may be termed the "cause," $c(t)$. We shall assume that $c(t)$ is always zero prior to $t = 0$, but that it is otherwise unrestricted in form (except by very liberal mathematical requirements always met in practical cases). The response which $c(t)$ evokes in some part of the circuit may be termed the "effect," $e(t)$. The effect will often involve a part called the "steady-state response," as well as a part which gradually dies away.

The classical method of transient analysis is to secure a general solution of the differential equations of the network, and to evaluate the arbitrary constants in it using conditions prevailing at time $t = 0$ (the initial conditions). This method is often very laborious in comparison with the Laplace method which will now be described (*1*).

The Laplace method is based on the functional transformation

$$F(s) = \int_0^\infty f(t)e^{-st}dt \qquad (1)$$

where the function of time $f(t)$ is zero for $t < 0$, and the variable $s = \sigma + j\omega$ is complex, having a real part σ sufficiently positive to insure the absolute convergence of the infinite integral. For any function of time $f(t)$, Eq. (1) defines uniquely a function $F(s)$, the *direct* transform of $f(t)$. Conversely, it can be shown that any $F(s)$ corresponds uniquely to a function of time $f(t)$ which will satisfy Eq. (1).* The function $f(t)$ is termed the *inverse* transform of $F(s)$.

One important advantage of the transformation (1) is that complicated discontinuous functions which may consist of pieces of functions of different analytic form lead to simple continuous functions $F(s)$. As a first example, consider the unit step function $u(t)$ defined as follows:

$u(t) = 0, \quad t < 0; \quad u(t) = 1, \quad t \geq 0.$

The direct transform is

$$u(s) = \int_0^\infty e^{-st}dt = \frac{1}{s}, \qquad \sigma \geq c > 0.$$

* There are certain matters regarding the mathematical behavior of $f(t)$ at discontinuities which we are omitting.

Table 1

Some Laplace Transform Pairs

$f(t)$	$F(s)$
$af_1(t) + bf_2(t)$	$aF_1(s) + bF_2(s)$
$df(t)/dt$	$sF(s) - f(+0)$†
$\int f(t)dt$	$F(s)/s + f^{(-1)}(+0)/s$
$e^{-at}f(t)$	$F(s + a)$
$f(t - a)$ $[f(t - a) = 0, -\infty < t < a]$	$e^{-as}F(s)$
$u_1(t)$ (unit impulse at $t = 0$)	1
$u_2(t)$ (unit doublet at $t = 0$)	s
$u(t)$ (unit step at $t = 0$)	$1/s$
$t^{n-1}/(n - 1)!$	$1/s^n$ $n = 2, 3, \cdots$
e^{-at}	$1/(s + a)$
$(1/b) \sin bt$	$1/(s^2 + b^2)$
$\cos bt$	$s/(s^2 + b^2)$
$(1/b)e^{-at} \sin bt$	$1/[(s + a)^2 + b^2]$

† $f(+0)$ is the value of $f(t)$ at $t = 0$. The + sign indicates that in the case of a discontinuity at $t = 0$, $f(+0)$ is value approached from the positive side.

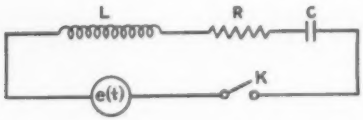


FIG. 1. Series circuit used to illustrate method of Laplace transformation

As a second example, consider the function

$f(t) = 0, \quad t < 0; \quad f(t) = e^{-at}, \quad t \geq 0.$

Here

$$F(s) = \int_0^\infty e^{-(a+s)t}dt = \frac{1}{s+a},$$

$$\sigma \geq c > \mathrm{Re}(a).$$

Tables connecting $F(s)$ with various functions $f(t)$ are found in standard texts and can be used to find $F(s)$ for a given $f(t)$, or *vice versa*. Some of the most important transforms which one needs are contained in Table 1. It will be noted that some of the pairs give the transforms of operations.

The method by which the Laplace transformation is used to obtain the transient response of a network is presented most simply by treating a particular case. The simple series network shown in Fig. 1 contains the circuit elements most commonly occurring.

It is desired to find the current $i(t)$ which flows in the circuit of Fig. 1 when the key K is closed at zero time, connecting a given source of voltage $e(t)$ to the formerly quiescent circuit. The differential equation of the network is

$$\mathrm{L}\frac{di}{dt} + \mathrm{R}i + \frac{\int idt}{\mathrm{C}} = e(t) \qquad (2)$$

We first obtain the Laplace transform of Eq. (2) by multiplying each term by $e^{-st}dt$ and integrating from 0 to infinity. Using Table 1, we find that

$$\mathrm{L}sI(s) + \mathrm{R}I(s) + I(s)/\mathrm{C}s = E(s) \qquad (3)$$

where we have used the fact that $i_0 = q_0 = 0$ in obtaining from the table the transform of the derivative and the indefinite integral of $i(t)$. Equation (3) is now solved by algebra giving for the transform of the current

$$I(s) = \frac{E(s)}{\left(\mathrm{L}s + \mathrm{R} + \frac{1}{\mathrm{C}s}\right)} \qquad (4)$$

The current $i(t)$ is the inverse transform of $I(s)$, and can be evaluated from a table of Laplace transforms, or by direct means which cannot be described here, as soon as $e(t)$ is specified so that $E(s)$ is known. For instance, if $e(t)$ is a battery of emf E_0, then $e(t) = E_0u(t)$, and $E(s) = \frac{E_0}{s}$. In this case

$$I(s) = \frac{E_0}{\mathrm{L}s^2 + \mathrm{R}s + \frac{1}{\mathrm{C}}}$$

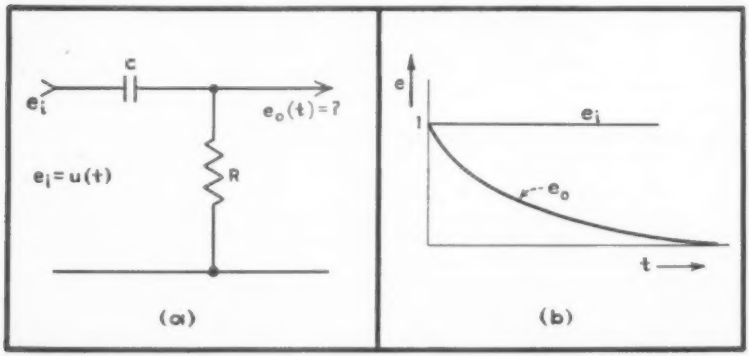


FIG. 2. Simple RC coupling network and its response to a step-function voltage signal

$$= \frac{E_0}{L} \frac{1}{\left(s + \frac{R}{2L}\right)^2 + \frac{1}{LC} - \frac{R^2}{4L^2}}$$

with the familiar result that

$$i(t) = \frac{E_0}{L\sqrt{\frac{1}{LC} - \frac{R^2}{4L^2}}} \exp\left(-\frac{R}{2L}t\right) \sin\sqrt{\frac{1}{LC} - \frac{R^2}{4L^2}}\, t \quad (5)$$

The method used in the example is quite general and can be applied to the solution of any linear differential equation with constant coefficients for assigned initial conditions. The method becomes more useful in the case of electrical networks on noting that the denominator of the right-hand member of Eq. (4) is simply the complex impedance of the network, with the usual $j\omega$ replaced by the complex variable s. The transient response of a quiescent network can be obtained, therefore, from the sinusoidal steady-state response, using the following procedure:

1. First obtain the steady-state response using complex impedances and replace each $j\omega$ by s, obtaining in this way the system function $G(s)$.

[In the above example

$$G(s) = \frac{1}{Ls + R + \frac{1}{Cs}}$$

]

2. Obtain the transform of the cause

$$C(s) = \int_0^\infty c(t)e^{-st}dt = \mathfrak{L}[c(t)]$$

3. Obtain the transform of the effect as follows:

$$E(s) = G(s) \cdot C(s)$$

4. Obtain the desired transient response by finding the inverse transform of $E(s)$, using tables, or other means. As a formal process this may be written

$$e(t) = \mathfrak{L}^{-1}[E(s)].$$

Let us now consider a number of examples which are very often met in electronic circuits.

II. Response of Various Circuit Elements to a Step Function

1. "Differentiation" of a step function. A step function input signal is applied to the simple RC network shown in Fig. 2*a*, and it is desired to find the form of signal occurring across the resistance R. This network is often used in coupling together two points in a circuit having different d-c potentials. It is also used in shaping pulses in a pulse amplifier.

Here the system function takes the form

$$G(s) = \frac{R}{R + \frac{1}{Cs}} = \frac{\tau s}{1 + \tau s}$$

where $\tau = RC$. The transform of the output voltage is

$$E_0(s) = \frac{1}{s}G(s) = \frac{\tau}{1 + \tau s}$$

which has the inverse transform

$$e_0(t) = \exp\left(-\frac{t}{\tau}\right).$$

The two voltage signals are shown in Fig. 2*b*.

2. Double "differentiation" of a step function. In most amplifiers, two or more RC coupling networks are necessary, one of which may be used for shaping pulses in the case of an amplifier designed to amplify pulses from an electrical detector of radiation. For two coupling networks, we are led to consider the equivalent circuit shown in Fig. 3*a*. We now have the system function

$$G(s) = \frac{\tau_1 s}{1 + \tau_1 s} \cdot \frac{\tau_2 s}{1 + \tau_2 s}$$

The transform of the output signal, in

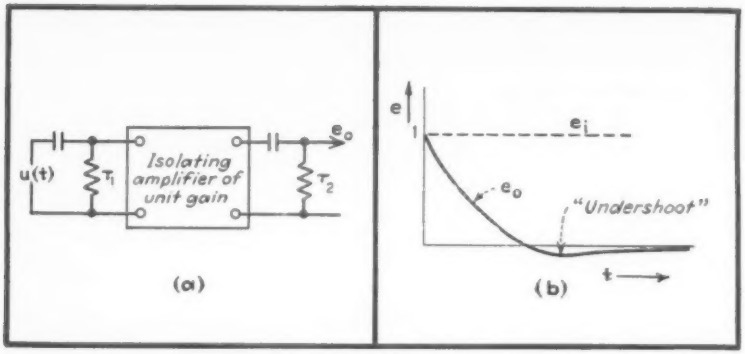


FIG. 3. Circuit and curve illustrating double differentiation of a step function

response to a step signal input, is

$$E_0(s) = \frac{1}{s} G(s),$$

from which it is found that

$$e_0(t) = \frac{1}{\tau_2 - \tau_1}\left[\tau_2 \exp\left(-\frac{t}{\tau_1}\right) - \tau_1 \exp\left(-\frac{t}{\tau_2}\right)\right].$$

Additional stages each contribute an "undershoot" or overshoot to the transient response. To avoid this in pulse-shaping applications, one time constant should be much smaller than the rest (by a factor of 100 for 1% undershoot).

3. Plate load of an R-coupled amplifier. By replacing the vacuum tube of a pentode amplifier stage by a constant-current generator, we are led to consider the transient problem indicated in Fig. 4*a*, where R is the plate load resistance and C the stray, or "parasitic," capacitance shunting it. For this case,

$$G(s) = Z(s) = \frac{R'}{1 + \tau s}; \qquad \tau = RC,$$

so that $E_0(s) = \frac{I_0}{s} \cdot G(s)$

The output voltage now takes the form

$$e_0(t) = I_0 R\left[1 - \exp\left(-\frac{t}{\tau}\right)\right]$$

which is shown in Fig. 4*b*.

The response rises from 10 to 90% in the time 2.2τ. This is the so-called *rise time* according to one definition.

4. Plate load of a shunt-compensated amplifier. A somewhat faster rise is obtained for given values of R and C if a small inductance is placed in series with R, a procedure known as *shunt compensation.* The system function of the equivalent circuit given in Fig. 5*a* takes the form

$$G(s) = Z(s) = \frac{1}{\frac{1}{R + Ls} + Cs} = \frac{R + Ls}{1 + RCs + LCs^2}.$$

Further analysis is simplified if we set R = 1, C = 1 so that $L = \alpha R^2 C = \alpha$. This device normalizes the system function (and the response to a unit step function), and establishes a time scale in units of RC. The normalized system function becomes

$$g(s) = \frac{1 + \alpha s}{1 + s + \alpha s^2},$$

giving the transform of the response to a unit step function current,

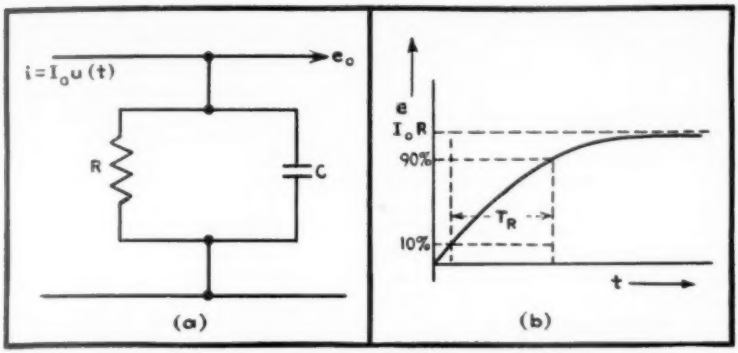


FIG. 4. Equivalent plate load of a resistance-coupled amplifier stage, and its response to a unit step function

$$E_0(s) = \frac{1}{s}g(s) = \frac{1}{s}\frac{s + \frac{1}{\alpha}}{(s + s_1)(s + s_2)},$$

where

$$s_1, s_2 = -\frac{1}{2\alpha} \pm \sqrt{\frac{1}{4\alpha^2} - \frac{1}{\alpha}}$$

Using the transform pair (1.109) in Gardner and Barnes (*1*), we find the transient response

$$e_0(t) = 1 - \frac{\left(\frac{1}{\alpha} - s_1\right)}{s_1(s_2 - s_1)} e^{-s_1 t} - \frac{\left(\frac{1}{\alpha} - s_2\right)}{s_2(s_1 - s_2)} e^{-s_2 t},$$

which can be reduced to other forms if desired. The response will be oscillatory if s_1 and s_2 have an imaginary part. For no oscillations to occur evidently $\alpha \leq 1/4$. If $\alpha = 1/4$, the network is said to be critically compensated, and for this case

$$E_0(s) = \frac{1}{s}\frac{(1 + 1/4\, s)}{(1 + 1/2\, s)^2}$$

which yields

$$e_0(t) = 1 - (1 + t)e^{-2t}$$

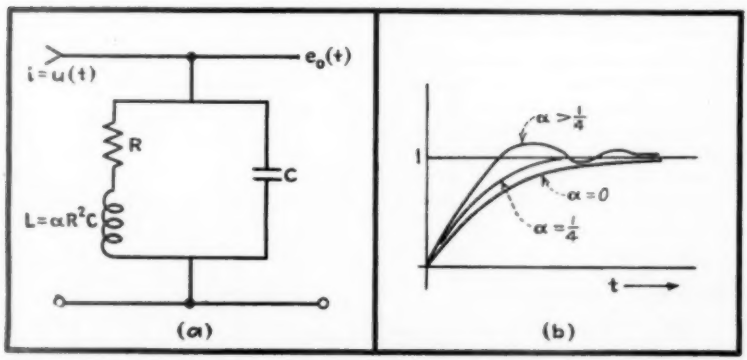


FIG. 5. Equivalent plate load of a shunt compensated amplifier stage, and its response to a unit step function

using the pair (2.138) in Gardner and Barnes.

The 10 to 90 rise time for the critically compensated case is 1.55τ, so the compensation has decreased the rise time by the factor $1.55/2.2 = 0.71$. If this method of compensation is used in a practical amplifier, it is convenient to make the inductance variable with a movable dust core, and to adjust it while examining the transient response with a cathode-ray oscilloscope. An indication of what happens when α is varied is shown in Fig. 5*b*.

It should be noted that in the present example the roots of the denominator of $G(s)$ must have no imaginary part if the response is to be non-oscillatory (or monotonic). This condition, however, is neither necessary nor sufficient to insure a monotonic response in the case of an arbitrary network.

5. Transient amplifiers (in general). Let $G(s)$ be the complex gain of a linear amplifier. If $E_i(s) = \mathcal{L}[e_i(t)]$ is the transform of the input signal, then the output signal is given by

$$e_0(t) = \mathcal{L}^{-1}[G(s) \cdot E_i(s)].$$

In studying the transient response of a low-pass amplifier, it is often convenient to split $G(s)$ into two factors, $G(s) = G_1(s) \cdot G_2(s)$, where the first factor is the system function of an equivalent amplifier idealized to have a flat frequency response characteristic at high frequencies, and the second factor is the system function of an equivalent amplifier idealized to have a flat characteristic at low frequencies (down to zero frequency). The transient response for short times, and for long times can then be treated independently. The gain-frequency curves corresponding to G_1 and G_2 are illustrated in Fig. 6.

TABLE 2

Relative Pulse Amplitudes

λ	$\lambda^{1/1-\lambda}$
0.5	.250
1.25	.410
2.5	.543
5.0	.669
12.5	.803
25.	.845
50.	.923
125.	.962
250.	.978
∞	1.000

6. Amplifier having a single RC cut-off at low and at high frequencies. The system function for the amplifier stage shown in Fig. 7 takes the form

$$G(s) = \frac{s\tau_1}{1 + \tau_1 s} \cdot \frac{R_2 g_m}{1 + \tau_2 s}$$

where $\tau_1 = R_1C_1$ and $\tau_2 = R_2C_2$. The lower and upper half-power frequencies of the frequency response characteristic are located at $f_1 = \frac{1}{2\pi\tau_1}$ and $f_2 = \frac{1}{2\pi\tau_2}$, respectively. The normalized system function is evidently

$$\frac{G(s)}{R_2 g_m} \equiv g(s) = \frac{s\tau_1}{(1 + \tau_1 s)(1 + \tau_2 s)}.$$

The transform of the response to a unit step function

$$E_0(s) = \frac{R_2 g_m}{s} g(s)$$

gives the transient response (a *pulse*)

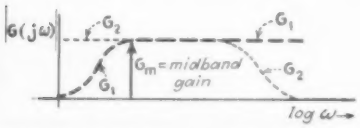


FIG. 6. Gain-frequency characteristics of a low-pass amplifier

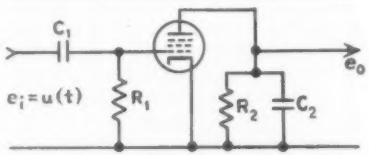


FIG. 7. Equivalent circuit of amplifier stage

$$e_0(t) = R_2 g_m \times \frac{\tau_1}{\tau_1 - \tau_2}\left[\exp\left(-\frac{t}{\tau_1}\right) - \exp\left(-\frac{t}{\tau_2}\right)\right]$$

By setting $de_0/dt = 0$, it is found that the pulse has the amplitude

$$e_{max} = R_2 g_m \cdot \lambda^{1/1-\lambda}$$

where $\lambda \equiv \tau_1/\tau_2$. Some values of the function $\lambda^{1/1-\lambda}$ are given in Table 2.

III. Theory of the Transient Response of Voltage Amplifiers

1. Equivalent circuits for a linearly operated triode or pentode. It is assumed that the reader is familiar with the characteristic curves of vacuum tubes, such as those given in the "RCA Tube Manual." Although these curves, together with accompanying numerical data, are invaluable in designing practical circuits, a mathematical analysis of amplifier transient response requires the use of an equivalent circuit for each vacuum tube, based on the assumption that the tube behaves linearly in the range of signals being considered.

For a triode, and for a pentode whose screen voltage is held fixed with respect to the cathode,

$$I_g = 0, \qquad I_p = I_p(E_g, E_p), \tag{6}$$

where I_g is the grid current, I_p is the plate current, and E_g and E_p the grid and plate potentials measured with respect to the cathode.

Letting $i_p = \Delta I_p$, $e_g = \Delta E_g$, etc., we have that

$$i_p = \left(\frac{\partial I_p}{\partial E_g}\right)_{E_p} e_g + \left(\frac{\partial I_p}{\partial E_p}\right)_{E_g} e_p = g_m e_g + \frac{e_p}{r_p} \tag{7}$$

where $g_m = \left(\frac{\partial I_p}{\partial E_g}\right)_{E_p}$ and $\frac{1}{r_p} = \left(\frac{\partial I_p}{\partial E_p}\right)_{E_g}$

g_m is the transconductance and r_p the plate resistance of the tube. For small changes in current and voltage, it is customary to assume g_m and r_p are constants, i.e., the tube operates in a linear fashion. If now a resistance R_L is placed in series with the plate connection,

$$e_p = -i_p R_L$$

so that Eq. (7) becomes

$$+\mu e_g = i_p r_p + i_p R_L \tag{8}$$

where $\mu = g_m r_p = -\left(\frac{\partial E_p}{\partial E_g}\right)_{I_p}$ is the

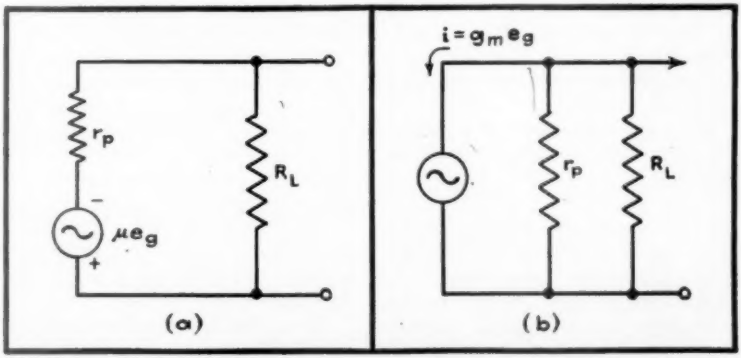


FIG. 8. Equivalent circuits for a triode or pentode; (*a*) constant-voltage generator, (*b*) constant-current generator

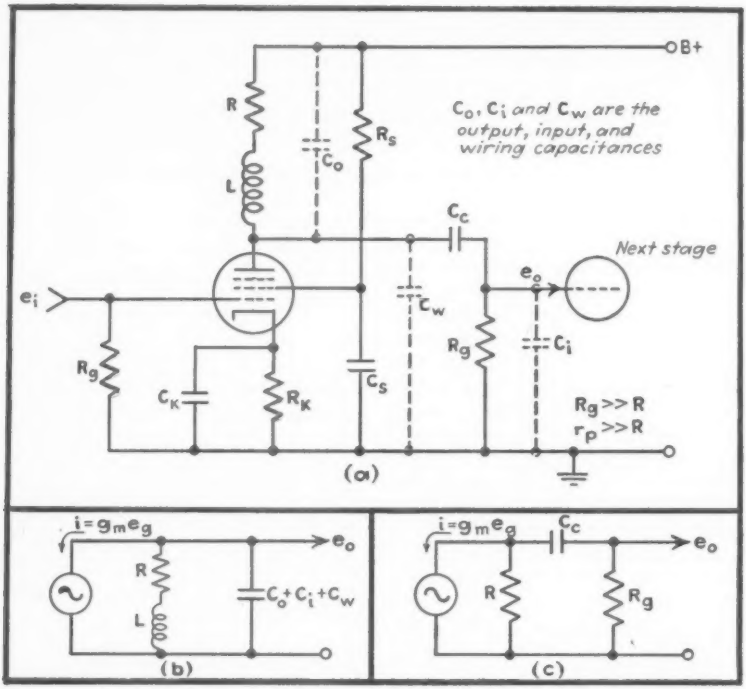


FIG. 9. (*a*) Typical amplifier stage; (*b*) equivalent circuit at high frequencies; (*c*) equivalent circuit at low frequencies

amplification factor. Two convenient circuits equivalent to Eq. (8) are given in Figs. 8*a* and 8*b*. The constant-current form is most convenient for a pentode, since $r_p \gg R_L$ in most applications of interest.

If the screen voltage E_s of a pentode can vary (with respect to the cathode), then

$$I_g = 0,$$
$$I_p = I_p(E_g, E_s, E_p),$$
$$I_s = I_s(E_g, E_s, E_p),$$

from which *six* independent tube parameters emerge. By the method used above, it is not difficult to obtain a complete equivalent circuit for this case. It will involve a plate and screen resistance, r_p and r_s, and four voltage (or current) generators.

In deriving the two sample equivalent circuits, we have neglected inter-electrode capacitances. Since the coupling between plate and grid of a triode results in a lower input impedance (the Miller effect), this type of tube ordinarily is not used in a wide-band (fast) amplifier. The shielding present in a pentode reduces the coupling to a point where it can be neglected, so the equivalent circuits given are sufficiently accurate, provided input and output capacitances are supplied.

The equivalent circuit for a complete amplifier stage usually can be drawn by inspection, as the example shown in Fig. 9 will illustrate. The screen and

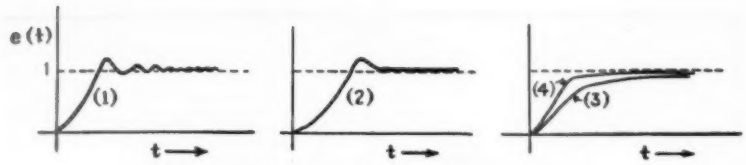


FIG. 10. Some typical transient response curves

cathode bias resistors are assumed to be bypassed adequately, and it is assumed that $R_g \gg R$ and $r_p \gg R$.

2. Transient response of equivalent high-frequency amplifier (*2*). Let $G_2(s)$ be the gain function of the equivalent high-frequency amplifier, which possesses perfect response at low frequencies. If $G_2(0)$ is the gain at zero frequency of the idealized amplifier, then

$$g_2(s) = \frac{G_2(s)}{G_2(0)} \qquad (9)$$

is the normalized gain function. Calculation of the transient response of the system function (9) to a unit step function $u(t)$ is the customary way of studying the transient properties of the amplifier for signals possessing high-frequency components (fast signals). This response is found from the inverse Laplace transform

$$e(t) = \mathfrak{L}^{-1}\left[\frac{1}{s}\, g_2(s)\right], \qquad (10)$$

and the time derivative of this response (or the response to a unit impulse) is

$$e'(t) = \mathfrak{L}^{-1}[g_2(s)]. \qquad (11)$$

The direct transform corresponding to Eq. (11) is

$$g_2(s) = \int_0^\infty e'(t)e^{-st}dt. \qquad (12)$$

Some typical transient response curves are indicated in Fig. 10. Curves (1) and (2) show overshoot of an oscillatory and non-oscillatory sort, respectively. Curves (3) and (4) are monotonic, with (4) reaching the final value in a minimum time consistent with the sort and number of tubes, and the total gain of the amplifier. Response curves (3) and (4) are considered more desirable than (1) and (2) for a transient amplifier. Subsequent discussion will be limited to amplifiers having a monotonic response.

For many purposes the delay time (T_D) and rise time (T_R) adequately characterize the high-frequency transient properties of an amplifier. The delay time is often defined as the time interval between the application of a step signal and the time at which the output response has reached the half-value point, as indicated in Fig. 11*a*. The rise time is then defined as the reciprocal of this slope at this point (see figure). Another definition of T_R is the time interval between 10 and 90 % response. These definitions are awkward for making computations and have prompted some new definitions which, however, apply only to the monotonic type of response. For the delay time we can define

$$T_D = \int_0^\infty te'(t)dt \qquad (13)$$

which is the centroid of $e'(t)$; and for the rise time

$$T_R^2 = 2\pi \int_0^\infty (t - T_D)^2 e'(t)dt$$
$$= 2\pi\left[\int_0^\infty t^2 e'(t)dt - T_D^2\right] \qquad (14)$$

which makes the rise time $\sqrt{2\pi}$ times the standard deviation of the curve $e'(t)$. The factor $\sqrt{2\pi}$ is chosen so as to make this value of rise time agree with the one based on slope when $e'(t)$ is a Gauss error curve, which is the limiting form $e'(t)$ takes when the number of amplifier stages increases

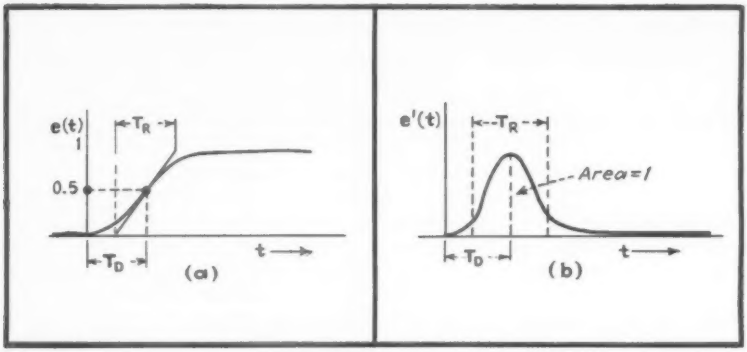


FIG. 11. Curves illustrating the definition of rise time and delay time

without limit. In practice values of T_D and T_R, from (13) and (14), will differ but little from the "laboratory" definitions in current use. The usefulness of (13) and (14) will now be demonstrated.

In Eq. (12) expand e^{-st} in an infinite power series, to obtain

$$g_2(s) = 1 - s\int_0^\infty te'(t)dt + \frac{s^2}{2}\int_0^\infty t^2e'(t) - \cdots$$

Hence if a given system function is expanded in powers of s, *i.e.*,

$$g_2(s) = 1 + As + Bs^2 + \cdots,$$

we have, by inspection, that

$$T_D = -A,$$

and that

$$T_R = \sqrt{2\pi}\,(2B - A^2)^{1/2}$$

It is convenient to consider system functions of the form

$$g_2(s) = \frac{1 + a_1s + a_2s^2 + \cdots}{1 + b_1s + b_2s^2 + \cdots},$$

since they often occur in practice. Here

$$\left.\begin{aligned} T_D &= b_1 - a_1, \\ \text{and} \\ T_R &= \sqrt{2\pi} \\ &\quad [b_1^2 - a_1^2 + 2(a_2 - b_2)]^{1/2} \end{aligned}\right\} \quad (15)$$

Let us now consider two examples, a resistance-coupled amplifier stage, and the corresponding shunt-compensated stage. Expressions for the transient response to a step function for both these cases have been derived in Sec. II.

The normalized system function of a resistance-coupled stage takes the form

$$g_2(s) = \frac{1}{1 + RCs}.$$

On computing the delay time and rise time using Eqs. (15), we find that

$$T_D = RC,$$

and $$T_R = \sqrt{2\pi}\,RC = 2.51RC,$$

that is, the delay time equals the plate circuit time constant, and the rise time is about two and one-half times this time constant.

The "slope" definition gives $T_R = 2RC$ and the "10-to-90" definition, $T_R = 2.2RC$.

The gain, rise-time quotient for the stage, computed from $G_0 = g_mR$ and $T_R = \sqrt{2\pi}\,RC$, is

$$\frac{G_0}{T_R} = \frac{g_mR}{\sqrt{2\pi}\,RC} = \frac{g_m}{\sqrt{2\pi}\,C} \quad (16)$$

Evidently the quantity $\frac{g_m}{\sqrt{2\pi}\,C}$ measures the excellence of the tube for amplifying fast transients.

For a 6AC7, $g_m = 0.009$ mhos, $C = 8 + 11 = 19$ $\mu\mu f$, which make $G_0/T_R = 190$ per microsecond. If one includes the stray wiring capacitance, then $C = 23$ $\mu\mu f$ approximately, and $G_0/T_R \doteq 155$ per microsecond. For a 6AK5, $g_m = 0.005$ mhos, $C = 4.3 + 2.1 = 6.4$ $\mu\mu f$, making $G_0/T_R = 312$ per microsecond. If again wiring capacitance is included, $C = 10.4$ $\mu\mu f$ approximately, so $G_0/T_R \doteq 208$ per microsecond.

The normalized system function of a shunt-compensated amplifier stage (see Fig. 5) may be written

$$g_2(s) = \frac{1 + \frac{1}{4}RCs}{1 + RCs + \frac{1}{4}R^2C^2s^2}$$

so that, by Eqs. (15),

$$T_D = \tfrac{3}{4}RC,$$

$$T_R = \sqrt{\tfrac{7}{16}}\sqrt{2\pi}\,RC = 1.66\,RC.$$

The 10-to-90 definition gives the value 1.5RC.

The gain, rise-time quotient is now

$$\frac{G_0}{T_R} = \sqrt{\frac{16}{7}}\frac{g_m}{\sqrt{2\pi}\,C}, \qquad (17)$$

For the general case, this quotient takes the form

$$\frac{G_0}{T_R} = S \cdot \frac{g_m}{\sqrt{2\pi}\,C} \qquad (18)$$

where S may be defined as the rise time figure-of-merit for the type of coupling used. Evidently $S = 1$ for an R-coupled stage, and $S = (1\frac{6}{7})^{1/2}$ for a shunt-compensated stage. The theoretical limit for S appears to be $3/\sqrt{2} = 2.12$, for two-terminal interstages, compensated for no overshoot in the transient response to a step function.

Let us next consider some theorems which apply to multi-stage amplifiers where the gain function of the entire amplifier is the product of the gain functions of the individual stages. (No feedback interactions.) The proof of these theorems is based mostly on the definitions (13) and (14), and may be found in reference (*1*).

(A)

$$\left.\begin{aligned} T_D &= \sum_1^n T_{Di}, \\ T_R^2 &= \sum_1^n T_{Ri}^2, \end{aligned}\right\} \qquad (19)$$

i.e., delay times add, whereas the squares of rise times add.

(B) For a given gain, the rise time T_R of an amplifier is shortest when all n stages have the same rise time T_{R1}. When this is true,

$$T_R = \sqrt{n}\,T_{R1} \qquad (20)$$

(C) The gain of each stage of an amplifier of rise time T_R must be [from Eqs. (18) and (20)]

$$G_1 = \frac{S}{\sqrt{n}}\left(\frac{g_m}{\sqrt{2\pi}\,C}\right)T_R, \qquad (21)$$

giving a total gain

$$G_t = G_1^n. \qquad (22)$$

(D) If G_t is specified, then the rise time depends on n as follows: [from Eqs. (21) and (22)]

$$T_R = \left(\frac{\sqrt{2\pi}\,C}{g_m}\frac{1}{S}\right) \times \sqrt{n}\,G_t^{\frac{1}{n}} \qquad (23)$$

which leads to a minimum rise time when $n = 2 \ln G_t$ making

$$G_1 = e^{1/2} = 1.65 \cdots.$$

For instance, if $G_t = 10^5$, then $n \approx 23$ stages and the rise time will be greater for fewer or more stages, assuming the total gain to be held constant. The minimum rise time obtainable when $G_1 = e^{1/2}$ is

$$T_{min} = \frac{1}{S}\left(\frac{\sqrt{2\pi}\,C}{g_m}\right)\sqrt{2e \ln G_t}. \qquad (24)$$

Let us now compute the minimum rise time which can be obtained with ordinary tubes in a shunt-compensated amplifier. Let $S = 1.5$, $C = 22 \times 10^{-12}$, $g_m = .009$, $G_t = 10^5$ (a 23-stage, shunt compensated amplifier based on 6AC7's, having a voltage gain of 10^5). Equation (24) thus becomes

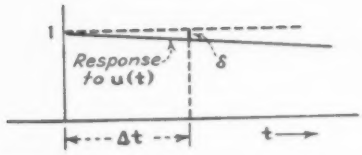


FIG. 12. Transient response curve illustrating the definition of δ

$$T_{min} = \frac{1}{1.5}\left(\frac{2.5 \times 22 \times 10^{-12}}{.009}\right)\sqrt{\frac{2 \times 2.72 \times 5}{.434}}$$

or, $T_{min} = .032$ μsec.

In such an amplifier, the plate load resistors would have a value of only 180 ohms, limiting the range of linear operation to less than one volt.

A more reasonable amplifier might contain nine stages. The rise time computed from Eq. (23) is then found to be 0.044 microseconds. The plate load resistors would have the value 400 ohms, limiting the range of linear operation to a few volts. By using a 6AG7 and a 6V6 for the last two stages, and sacrificing some of the fast rise, a shunt-compensated amplifier with reasonable linearity up to 40 volts (positive pulses), and with a rise time of 0.06 microseconds can be built. By adopting a four-terminal type of interstage in the amplifier (which will not be discussed here), the rise time of this amplifier can be reduced to perhaps 0.035 microseconds, without sacrificing the monotonic sort of transient response. Such interstages used with the 23-stage amplifier would yield a rise time of about 0.02 microseconds. A faster amplifier can be built by substituting 6AK5's for most of the 6AC7's.

In concluding the present section on the transient response of the equivalent high-frequency amplifier, it is worth mentioning the connection between rise time and the upper half-power frequency (the frequency at which the sinusoidal voltage gain has fallen to 70%, or the power gain to 50%) of the amplifier.

It can be shown that the transient response $e'(t)$ (to a unit impulse) of an n-stage amplifier approaches more and more the form of a Gauss error curve as n is increased (assuming the transient response $e_1(t)$ of each stage is monotonic). In a similar fashion, the gain-frequency curve approaches a Gaussian as n increases. The equation of the Gaussian frequency response giving a rise time T_R is

$$|g_2(j\omega)| = \exp(-\pi T_R^2 f^2).$$

On equating this gain function to $1/\sqrt{2}$, one finds that

$$T_R f_2 = \sqrt{\frac{\ln 2}{2\pi}} = \frac{1}{3.015} = \frac{1}{3} \quad (25)$$

where f_2 is the upper half-power frequency. It is found experimentally that this relationship holds very closely for any amplifier likely to be encountered in amplifying pulses from ion chambers, so the relationship is a useful one to remember. For instance, the upper half-power frequency of an amplifier having a 0.1 μsec rise time is 3.3 Mc per sec. The amplifier, of course, is assumed to possess a monotonic transient response to a step function.

3. Transient response of equivalent low-frequency amplifier. Let the normalized gain function be $g_1(s)$. The transient response of $g_1(s)$ to the unit step function $u(t)$ has the value unity for $t = +0$ and then decays towards zero with increasing time. If the amplifier is not to distort an arbitrary transient lasting a time Δt, then it can be required that the response to a unit step shall deviate from unity in this interval by less than some specified amount, expressed by the small fraction δ. Fig. 12 illustrates this situation.

The most convenient way of analyzing the transient behavior is to expand $g_1(s)$ in a power series in $(1/s)$,

$$g_1(s) = 1 + \frac{a_1}{s} + \frac{a_2}{s^2} + \cdots . \quad (26)$$

where the leading term is unity, since $g_1(s)$ has been normalized. Then, for small values of t (i.e., values of the order of Δt) the response to $u(t)$,

$$e(t) = \mathcal{L}^{-1}\left[\frac{1}{s}\, g_1(s)\right]$$
$$= \mathcal{L}^{-1}\left[\frac{1}{s} + \frac{a_1}{s^2} + \frac{a_2}{s^3} + \cdots\right]$$
$$= 1 + a_1 t + a_2 \frac{t^2}{2!} + \cdots , \quad (27)$$

is a rapidly converging power series in t whose second term accounts for the initial linear dropping off of the transient response.

To illustrate this method of analysis, suppose the amplifier contains n interstage couplings of the type shown in Fig. 13. Then

$$g_1(s) = \left(\frac{\tau s}{1 + \tau s}\right)^n$$
$$= 1 - \frac{n}{\tau s} + \frac{n(n+1)}{2!\tau^2 s^2} \cdots$$

so that

$$e(t) = 1 - n\frac{t}{\tau} + \frac{n(n+1)}{2 \cdot 2!}\left(\frac{t}{\tau}\right)^2 - \cdots$$

Evidently the response falls off initially at the rate n/τ, and τ, the time constant of the coupling networks, must be chosen to satisfy.

$$\delta = n\frac{\Delta t}{\tau}. \quad (28)$$

As an example, suppose that $\delta = .01$ (one percent distortion), for signals lasting $\Delta t = 100$ μsec, and that the amplifier contains 5 couplings ($n = 5$). Then

$$\tau = \frac{5 \times 10^{-4}}{.01} = .05 \text{ seconds},$$

e.g., R = 1 megohm and C = 0.05 μf would be satisfactory. A calculation of the lower half-power frequency (frequency at which the voltage gain is down 3 db, or 30%) indicates that it is

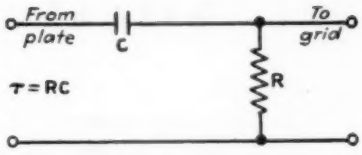


FIG. 13. Coupling network responsible for falling off of transient response

8.3 cps. If the amplifier shall amplify signals having a rise time of perhaps 0.5 μsec and lasting 100 μsec, with only a one-percent distortion due to time constants in the amplifier, the lower and upper half-power frequencies would have to be 8.3 cps and about 3 megacycles per sec respectively. The demands placed on the amplifier sinusoidal response are very severe indeed.

It is possible to incorporate compensating RC networks in the amplifier to make the coefficient a_1 in Eq. (26) or (27) equal to zero. This is normally done in video amplifiers, although the present method of analysis does not appear to be used in the literature for determining optimum values for elements in the networks employed (*3*). The tradition of analyzing amplifier behavior in terms of frequency response is somewhat unfortunate, unless one is primarily interested in amplifying steady sinusoidal signals.

In the case of amplifiers used in counting pulses from an electrical detector of radiation, the low-frequency response characteristic is deliberately modified in order to produce short pulses from the small voltage steps produced across the capacitance of the detector when each ionizing particle enters the detector.

BIBLIOGRAPHY

1. M. F. Gardner, J. L. Barnes, "Transients in Linear Systems," (John Wiley and Sons, Inc., New York, N. Y., 1942)
2. W. C. Elmore, *J. Applied Phys.* (in press)
3. F. E. Terman, "Radio Engineers' Handbook," p. 414–418 (McGraw-Hill Book Co., New York, N. Y., 1943)

OXYGEN-18 AS A TRACER ELEMENT

A critical review of the characterization, preparation, and techniques for application of oxygen-18. Its uses to study metabolic, organic, and inorganic reactions are analyzed in detail and supplemented by an extensive bibliography

By RONALD BENTLEY*

Commonwealth Fund Fellow, Department of Biochemistry
College of Physicians and Surgeons, Columbia University, New York

THE POSSIBILITY THAT OXYGEN was not a simple element was first suggested by Blackett in 1925. In his study of the ejection of protons from nitrogen nuclei upon bombardment with fast alpha particles, a number of anomalous cloud-chamber tracks were observed. It was deduced that these were probably due to a nucleus of mass seventeen and atomic number eight. Blackett concluded that this nucleus was an oxygen isotope which, if stable, would exist on the earth, but in such small quantities as to escape detection in the mass spectrograph of Aston.

Some years later, Giauque and Johnston (*1*) showed that the weak doublets in the atmospheric absorption bands of oxygen, observed earlier by Dieke and Babcock, could be interpreted on the quantum theory as due to a molecule $O^{18}O^{16}$. The calculated shifts, attributed to the vibrational and rotational isotope effects, agreed well with the observed separations. Later, spectrograms at low solar altitudes decisively confirmed the existence of O^{18} (*2*). These plates also showed a number of extremely weak lines, and Giauque and Johnston were able to demonstrate that these were due to the molecule $O^{17}O^{18}$. Aston at this time, using the mass spectrograph, was unable to obtain evidence for either isotope. Later, however, Ruckärdt confirmed O^{18} by electromagnetic analysis of canal rays (*3*).

The early values for the relative abundances of the isotopes were somewhat erratic, but later work by Bleakney and Hipple with a mass spectrometer gave the ratio $O^{16}:O^{18}$ for a large number of samples at around 500. Murphey (*4*), using Nier's precision spectrometer, gave the ratio $O^{16}:O^{18}$ as 500 ± 15 and $O^{18}:O^{17}$ as 4.9 ± 0.2. The figures in use today are:

Mass Number	16	17	18
% Abundance	99.76	0.04	0.20

On the basis of data obtained in the mass spectrograph, the isotopic weights of O^{18} and O^{17}, respectively, are 18.00485 ± 0.00018 and 17.00450 ± 0.00006.

Attempts were soon made to enrich the heavy isotopes. It was found by Washburn *et al.* (*5*) that the isotopic fractionation taking place during the electrolysis of water resulted from concentration of the heavy isotopes in the residual water. It was later pointed out that the initial failure to achieve any fractionation could be explained as resulting from carbonation of the concentrated alkali solutions obtained in

* Present address: National Institute for Medical Research, Hampstead, London, N.W. 3.

the electrolytic process with carbon dioxide—a process which results in "equilibration" of the water to almost the normal isotopic abundance (*6*). The efficiency of the electrolytic process may be gaged from the fact that by the electrolysis of 117 liters of commercial electrolyte to 1 ml, O^{18} concentration increased only from 0.202% to 0.222%.

Lewis and Cornish in 1933 showed that a slight difference in vapor pressure existed between the two forms of water H_2O^{16} and H_2O^{18}. Stedman (*7*) took advantage of this in achieving an appreciable O^{18} concentration by the fractionation of water in a crumpled stainless-steel column. This method, investigated intensively by Urey and his co-workers, finally resulted in a fractionation column that produced several hundred ml of heavy water, of maximum O^{18} concentration 0.85% (*8*). The column was 35 feet long, and consisted of alternate stationary and rotating cones which provided a large surface for reaction and at the same time offered only a small working hold-up. In a later experiment the same column was used to prepare a heavy methanol; the O^{18} content was doubled after several days (*9*).

The operation of a cascade of three 25-foot columns packed with Haydite aggregate was described by Thode *et al.* in 1944 (*10*). After operation for 120 days, it produced 150 ml of water enriched 6.5 fold with respect to O^{18} and 2.7 fold with respect to O^{17}. In all, 23 gm of O^{18} in excess of normal were produced.

Concentration of O^{18} has similarily been observed on evaporation of liquid oxygen to a small volume; oxygen obtained by fractionation of liquid air also contains about 2 in 10^5 more atoms of O^{18} than ordinary air.

Lewis observed in 1933, a simple type of isotopic reaction in which O^{18} atoms were exchanged as follows (*11*):

$$SO_2^{16} + H_2O^{18} = SO_2^{18} + H_2O^{16}$$

Later, the equilibrium constants and enrichment factors for a number of similar exchange reactions were calculated. In the case of carbon dioxide and water, it was found that the oxygen is concentrated in the former.

$$2H_2O^{18}\ (l) + CO_2^{16}\ (g) = 2H_2O^{16}\ (l) + CO_2^{18}\ (g)$$

The fractionation factor is 1.039 at 298.1° A. It was suggested that a fractionation column could be used for the reaction: The carbon dioxide collecting at the top would be hydrogenated to water and methane; the water would then be returned to the column, and a counter current process constructed. This process has not been used extensively for the enrichment of O^{18}.

The most complete separation of the oxygen isotopes ever achieved was that of Clusius, Dickel and Becker (*12*) by use of thermal diffusion. The equilibrium:

$$O_2^{16} + O_2^{18} = 2O^{16}O^{18}$$

was established over a hot platinum wire in 6 water-cooled units of total length 82 m. After operation for 18 months, 250 ml of O_2^{18} of 99% purity (1% $O^{17}O^{18}$) were obtained.

A partial separation of the isotopes by this method has also been recorded, and may be of value in the future. With six 3-m columns, O^{18} was enriched from the normal value to 17%. Still more recently, a unique process employing a combination of thermal diffusion and an exchange reaction has been described (*13*). The column establishes a counter current flow of two gases, which exchanges both C^{13} and O^{18} as follows:

$$C^{13}O + C^{12}O_2 = C^{12}O + C^{13}O_2$$
$$CO^{18} + CO_2^{16} = CO^{16} + CO_2^{18}$$

An 18% enrichment of O^{18} was obtained in 2.2 days; the corresponding enrichment of C^{13} is 23.5%.

It should be pointed out here that the O^{18}:O^{16} ratio shows slight variations

depending upon the source of the oxygen. Oxygen in fresh water has the same O^{18} content as oxygen in rocks that do not contain carbonates. Sea water has a slightly higher O^{18} content than fresh water, probably because of isotopic fractionation during evaporation. The O^{18} content of carbonate rocks is higher than that of fresh water. Carbonates are deposited from carbon dioxide in sea water which is in equilibrium with atmospheric carbon dioxide. This will consequently be enriched by the exchange reaction (previously discussed) with water for which the enrichment factor is 1.039 at 25°. Water prepared from atmospheric oxygen also shows an increased O^{18} content over that of fresh water. This so-called "Dole Effect" is discussed later in connection with photosynthesis.

Analytical Methods

No general method for the analysis of O^{18} in inorganic and organic compounds has as yet been devised, although it seems likely that one for organic compounds could be quickly developed (comparable to those used for deuterium and nitrogen-15). Such a process is discussed later.

The actual method to be used will be conditioned by the nature of the experiment and the analytical facilities available. In general, the analytical procedures involve either the use of a mass spectrometer or a measurement of the density of a water sample. The density determinations on even very small samples of water have been described for deuterium analyses and will be mentioned only briefly. The use of a mass spectrometer has been rather limited in the past; but commercial models are now available and extensive development work is being carried out on the design of a mass spectrometer for routine use.

The procedure in experiments involving inorganic compounds has usually been to observe the concentration changes of the aqueous solvent. Aten and Hevesy (*14*), however, analyzed heavy sodium sulphate by initial reduction with purified carbon at 900°, followed by reduction of the gases with hydrogen over a nickel catalyst to give water. This method should be capable of extension.

If oxygen gas can be obtained from a sample it may be directly analysed in the mass spectrometer. The atom % excess over normal may be calculated, if this method is used, from a determination of the mass ratios 32:33 and 32:34. In a number of cases, carbon dioxide may be readily obtained during the investigation, and this may be similarly analyzed directly in a mass spectrometer. The mass ratio 46:44 (designated as R) is determined, and the abundance is usually expressed as atom % excess by the following method.

In the gas sample, the following equilibrium will prevail:

$$CO_2^{16} + CO_2^{18} = 2CO^{16}O^{18}$$

Let mole fraction $CO^{16}O^{16} = a$

Let mole fraction $CO^{16}O^{18} = b$

Let mole fraction $CO^{18}O^{18} = c$

The measured abundance ratio

$$= \frac{CO^{16}O^{18}}{CO^{16}O^{16}} = \frac{a}{b} = R$$

By definition, atom % O^{18}

$$= \frac{b + 2c}{2(a + b + c)} \times 100 \quad (1)$$

The value of c may be expressed in terms of a and b, knowing the equilibrium constant of the above exchange reaction. This was determined by Urey and Greiff (1935) as 4.0 at 25°.

Therefore, $b^2/ac = 4$ and, hence, $c = b^2/4a$; by substitution in equation 1,

$$\text{atom \% } O^{18} = \frac{b}{2a + b} \times 100 = \frac{100}{2R + 1}$$

Subtraction of the normal abundance of O^{18} (0.20%) from this value then gives the atom % excess of O^{18}. The method is of particular interest in the

case of some carboxylic acids. Their metal salts are easily obtained in a high state of purity, and in general are degraded to carbon dioxide by heating in vacuum (e.g., lithium and silver salts). It would be also possible to determine the isotopic concentration of O^{18} by the similar use of gases such as carbon monoxide and sulphur dioxide, which could conceivably be obtained in some special cases.

The most nearly general method for the analysis of O^{18} in organic compounds would be to obtain the oxygen as water by the ter Meulen method (as used for nonisotopic oxygen analysis) whereby all of the oxygen atoms of an organic compound may be converted to water. [For an excellent review, see Russell and Fulton (*15*).] By this method, the sample to be analyzed is vaporized and then passed in a dry, oxygen-free, hydrogen stream over a red-hot cracking surface of platinum-coated quartz granules. The carbon dioxide, water, and hydrocarbon mixture so formed is next reduced by passing it over a very active thoria-promoted nickel catalyst, and the water formed frozen in a suitable trap. A quartz tube is used for the cracking and reduction process.

The method has been applied to methanol (first rectified and carefully dried) by Ingold and his co-workers (*16*). It should be quite feasible to carry out the operations on a micro or a semi-micro scale, analyzing the water sample by the equilibration method to be discussed later. A conversion apparatus could be readily devised in which all three operations, cracking, reduction, and equilibration, could be carried out concurrently.

Methanol has also been analyzed by passing the vapor in a hydrogen stream, first over a nickel spiral at 650°, and then over a thoriated nickel catalyst at 400°, trapping the water in a freezing mixture. A blank experiment showed that there was no isotope exchange between methanol and water under such conditions (*17*). Acetaldehyde was decomposed at 600° forming mainly methane and carbon monoxide; this mixture was similarly reduced by hydrogen over nickel at 270° (*18*).

Amyl alcohol has been directly dehydrated by passing repeatedly over bauxite at 400°. The pentane formed was pumped off, and the water purified for density determination by passing repeatedly over a platinum wire at 1200° (*19*).

For density determinations the water must be first vigorously purified and then electrolyzed, and the oxygen recombined with normal hydrogen to prevent any errors due to deuterium concentration. [See Datta, Day and Ingold, (*20*).] The following methods have been used in density determinations of H_2O^{18}.

The pressure float apparatus of the Gilfillan type: A pyrex glass float is totally immersed in the sample which is held at a constant temperature. By altering the hydrostatic pressure, the buoyancy of the float is varied, and the results obtained are compared with those using a standard sample. The maximum density measured by Cohn and Urey (*21*) using this method was 20 ppm, with an accuracy of 1 ppm.

The silica float method: The temperature at which a slim cylindrical silica float, completely immersed in the water sample, remains exactly suspended, is determined. A second observation with the same float in a standard liquid then gives a direct comparison of densities (*22*).

Drop methods: Aten and Hevesy (*14*) used the Linderstrøm-Lang floating drop method, and Mills, the falling-drop method with the micro-pipettes of Keston, Rittenberg and Schoenheimer (*23*). For a full description of the method, the article in "Preparation and

Measurement of Isotopic Tracers" (published by J. W. Edwards, Ann Arbor, Michigan) should be consulted.

As previously indicated, the most convenient method for the analysis of water containing O^{18} is to use the equilibration reaction with carbon dioxide. This exchange reaction leads to concentration of the O^{18} in gaseous carbon dioxide; the use of this method as an analytical tool came about as a result of the work of Cohn and Urey (21).

The procedure followed is to mix a known weight of water with carbon dioxide at atmospheric pressure, and to allow it to stand for some hours so that the equilibrium

$$CO_2^{16} + H_2O^{18} = CO^{16}O^{18} + H_2O^{16}$$

is completely attained. The water is then frozen, and the sample of equilibrated carbon dioxide is admitted to a mass spectrometer. The mass ratio 44:46 is determined in the usual way, and designated as R. The atom % excess O^{18} in the water may then be calculated as follows:

At equilibrium, let

number of moles $H_2O^{18} = a$
number of moles $H_2O^{16} = b$
number of moles $CO_2^{16} = c$
number of moles $CO^{16}O^{18} = d$
total number of moles of water $= x$
total number of moles of carbon dioxide $= y$
equilibrium constant $= K$

By consideration of the equilibrium reaction, it is now possible to set up the following equations:

$$\frac{bd}{ac} = K \qquad (2)$$

$$a + b = x \qquad (3)$$

$$c + d = y \qquad (4)$$

Atom fraction O^{18} in $CO^{16}O^{18}$

$$= \frac{d}{2c + 2d} = \frac{1}{R} \qquad (5)$$

Atom fraction O^{18} in $H_2O^{18} = \frac{a}{a+b}$ (6)

By elimination of the various unknowns, it can be shown that

Atom fraction O^{18} in H_2O^{18}

$$= \frac{1}{\frac{K}{2}(2R - 1) + 1}.$$

Therefore, atom % excess O^{18} in

$$H_2O^{18} = \frac{100}{\frac{K}{2}(2R - 1) + 1} - 0.20$$

The equilibrium constant K has been calculated as 2.076 at 298° A (*31*); hence the atom % excess of the water may be calculated from a determination of the mass ratio in the equilibrated carbon dioxide.

Only the stable isotopes of oxygen have been used in the study of chemical and biochemical processes. Two radioactive isotopes are known, O^{15} (positron emitter, half-life 124 ± 5 sec) and O^{19} (electron emitter, half-life 31 sec) but both are too short-lived to be of any practical use.

Heavy oxygen has been successfully applied as a tracer on many occasions, and now that limited quantities of H_2O^{18} (1.54 mole %) are available from the Atomic Energy Commission (*24*), it seems likely that its use will increase.

Before O^{18} can be used as a tracer in studying a particular reaction, it is first necessary to ascertain that simple exchange reactions are not taking place that will invalidate the results. A large number of both organic and inorganic compounds have been examined for exchange. Table 1 attempts to summarize the known exchange of inorganic compounds.

Many statements in the literature are conflicting because impure reagents were used in the experiments or there were present small amounts of material undergoing rapid exchange (e.g., in glass vessels, the metasilicate exchange is of considerable importance, particularly where a density determination is involved). In general it is necessary to have the salt in a definite and easily reproducible state of purity; it must be anhydrous and sufficiently soluble.

TABLE 1

Exchanges of Inorganic Compounds

Compound	*Extent of Exchange**	*Alkali effect*	*Acid effect*
H_3BO_3	Rapid		
$NaBO_2$	Complete in 4 hr		
$Na_2B_4O_7$	Rapid		
Na_2CO_3	Slow (half-exchange in 28 hr at 25)		
$KHCO_3$	Slow		
$NaNO_3$	None		
KNO_2	None		
Na_2SiO_3	Rapid		
K_3PO_4	None		
KH_2PO_4	Slow		
H_2SO_4	Complete		
Na_2SO_4	Very slow	Possibly catalyzed	
$KHSO_4$	Partial		
K_2SO_3	Complete	Complete inhibition	
$Na_2S_2O_3$	Complete	Complete inhibition	
$Na_2S_2O_5$	Partial at 19.3		
$NaClO_3$	None	Slight catalysis	Complete
$NaClO_4$	None	No effect	No effect
CrO_3	Complete at 19.3		
Na_2CrO_4	Complete at 70	Reduces exchange	
$Na_2Cr_2O_7$	Complete	Reduces exchange	
$KMnO_4$	Complete	No effect	
$KAsO_2$	Complete	No effect	
KH_2AsO_4	Complete	Slight catalysis	
Na_2SeO_3	Complete	No effect	
Na_2SeO_4	None	None	Partial
$NaBrO_3$	Slow	Slight catalysis	
K_2MoO_4	Complete		
$NaIO_3$	Slow	Slight catalysis	
Na_2WO_4	Complete		

*** All exchanges were made at 95–100° unless otherwise stated.**

These oxygen exchange reactions of inorganic salts provide useful information concerning the relative firmness of binding of oxygen atoms to the nonmetallic atom of the anion. The exchange reactions are very often catalyzed to a remarkable extent by acid or alkali; the only anions to have resisted exchange under any conditions are nitrite, nitrate, phosphate, sulphate, selenate, perchlorate, and chlorate. These are among the most weakly basic of the anions, and presumably contain oxygen atoms firmly bound to the nonmetal; however, some other anions of these atoms which are less weakly basic will exchange oxygen.

A second category of oxygen-containing anions comprises that group which shows only a partial or rather slow exchange, such as carbonate, bromate, iodate, chromate and permanganate. The exchange of bicarbonate has been shown to take place through the formation of carbonic acid and carbon dioxide, the rate of exchange depending upon the ratio of carbon dioxide to bicarbonate concentration. The time of half-exchange of 0.02 M sodium carbonate is 28 hours, the reac-

TABLE 2

Compound	*Temp. (°C)*	*Time (hours)*	*Catalyst*	*Extent of Exchange*
Methyl alcohol	100	7		None
	25	24	0.1*N* HCl	None
	25	48	0.1*N* NaOH	None
Glycerol	100	6		None
Phenol	25	44		None
	100	48		Slight
Acetic acid			HCl	Complete
			KOH	None
Monochloracetic acid	25	24		Partial
Trichloracetic acid	25	42		Complete
Butyric acid	100	24		None
Fumaric acid	25	44		None
	100	45		Partial
Maleic acid	100	45		Complete
Succinic acid	130	5		Complete
Benzoic acid	100	4		Complete
	100	4	0.1*N* HCl	Complete
Citric acid	25	24		None
	75	24		Complete
	25	24	H_2SO_4	Partial
Phthalic acid	100	20		Slight
Terephthalic acid	100	20		Slight
Amyl acetate	25	44		None
Acetaldehyde	25	24		Complete
Acetone	100	24		Partial
	25	1	0.002*N* HCl	Complete
	25	1	0.005*N* NaOH	Complete
Acetamide	25	71		None
Benzamide	100	23		None
Benzaldehyde	110	2		Complete
Urea	25	54		None
Glucose	100	48		Slight (1 atom exchanges)
Fructose, Galactose, Xylose, Arabinose	100	20		1 atom exchanges
Alloxan hydrate	25	430		87% exchange
Diphenylmethyl carbinol	95	10.5		None
	95	22	KOH	None
Trimethyl carbinol	95	20		None
	95	2	H_2SO_4	None
Trianisyl carbinol	95	43		None
	95	24	H_2SO_4	Complete
Glycine	100	24		None
Glycine hydrochloride (pH 1.9)	100	24		Complete
Tyrosine	100	40		None
Diketopiperazine	100	48		None
Glycylglycine	100	24		None

tion proceeding through the formation of the bicarbonate ion.

It is perhaps worth noting that the initial statement that phosphate underwent exchange has now been demonstrated to have been incorrect. When

sodium phosphate was dissolved in water enriched with respect to O^{18}, a density decrease of the water was observed. It was later pointed out that this density decrease was, in fact, due to an exchange with metasilicate derived from the glass container. When the experiment was repeated in a silver vessel, no such density decrease was observed.

Since the O^{18} in phosphate is stably bound, O^{18} could be used in biochemical studies on phosphate metabolism, providing a suitable analytical method can be devised. A high temperature dehydration or a modification of the method used by Aten and Hevesy for sulphate might be tried.

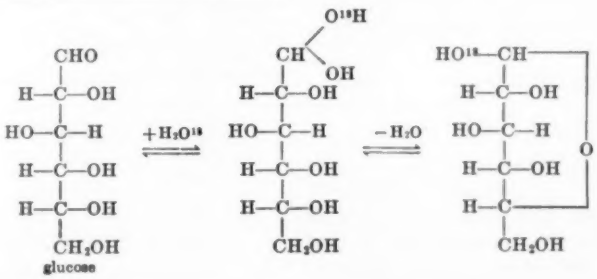
The exchange reactions of organic oxygen compounds have also been investigated, and it is possible to make the generalization that the oxygen of carbonyl and carboxyl groups is exchangeable. A list of the compounds which have been studied is given in Table 2.

The exchange of oxygen atoms in carboxyl groups is usually acid catalyzed, and when the acid is present as an anion, the exchange is usually completely inhibited. This effect of acid catalysis is illustrated by the series, acetic acid, monochloracetic acid and trichloracetic acid. With increasing acid strength from acetic to trichloracetic acid, the exchange takes place much more easily. The exchange with acetaldehyde is very rapid, the following equilibrium being established:

$$CH_3CHO + H_2O^{18} \rightleftharpoons CH_3CH(O^{18}H)(OH)$$

The similar, rather rapid exchange of benzaldehyde (even after rigorous purification to remove all traces of benzoic acid) is quite striking in view of the rather limited solubility of benzaldehyde in water. The existence of both acid and alkaline catalysis in the case of acetone provides further evidence of the amphoteric nature of the carbonyl group. It was concluded from a detailed kinetic study that the reaction does not proceed through the enolic form.

Alcoholic and phenolic hydroxyl groups do not usually exchange. However, in the case of the tertiary alcohol, trianisyl alcohol, it was possible to show an acid-catalyzed exchange, thus providing an interesting example of the labilization of hydroxyl groups by certain substituents. The sugars which have been investigated (glucose, fructose, galactose, xylose and arabinose) exchange only one oxygen atom, the exchange being complete in about twenty hours at 100°. The mechanism shown below, involving the opening of the oxide ring, was suggested:

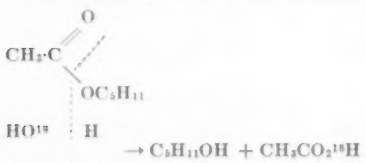


With glucose and fructose at room temperature, the rate of mutarotation is much more rapid than the rate of exchange of the oxygen atom. At higher temperatures, however, both processes seem to have the same rate.

Heavy oxygen exchange reactions of proteins have been studied in an attempt to obtain information regarding protein structure (*31*). Proteins contain a number of oxygenated functional groups, such as the peptide bond, alcoholic and phenolic hydroxyl groups, free carboxyl groups, amide groups, phosphate groups, and also water of hydration. Only carboxyl groups were found to exchange (e.g., in pepsin at pH 4.0) and no exchange was observed in the case of dry egg albumin. The observed exchange with pepsin was 13% and corresponded to the amount of oxygen present in the free carboxyl groups of the dicarboxylic amino acids contained in pepsin; the results confirmed current views concerning the amino acid distribution in pepsin.

Study of Ester Hydrolysis

The first use of O^{18} as a tracer in other than exchange reactions was the now classical study of ester hydrolysis carried out in 1934 by Polanyi and Szabo (*19*). Primary amyl acetate was subjected to alkaline hydrolysis in heavy water; the amyl alcohol obtained was dehydrated as described earlier. The experiment proved conclusively that the alkaline hydrolysis takes place as represented by the following equation:



$$\rightarrow C_5H_{11}OH + CH_3CO_2{}^{18}H$$

Somewhat later the case of acid hydrolysis was studied, and it was shown that when methyl hydrogen succinate was hydrolysed in H_2O^{18}, no O^{18} was found in the alcohol produced (*16*).

Roberts and Urey (*25*) pointed out the essential similarity of the mechanisms involved in ester hydrolysis, saponification (see later), and oxygen exchange: All are general acid-catalyzed and are first order in alcohol or water; they all proceed via the formation of similar active complexes, the disappearance of which controls the rate of reaction.

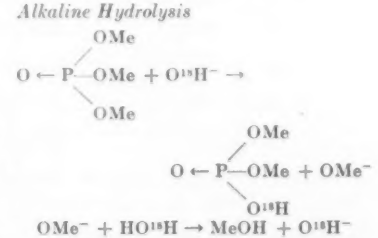
The study of the esterification of benzoic acid with heavy methanol showed that the water eliminated in the process did not contain any isotope (*17*). As it was known that benzoic acid exchanges only slowly, the mechanism of the reaction could be decided upon as:

$$C_6H_5CO\boxed{OH + H}O^{18}CH_3$$
$$\rightarrow C_6H_5CO^{18}OCH_3 + H_2O$$

The rate of the acid-catalyzed exchange reaction of benzoic acid was shown to be first order in the difference of the O^{18} content of the reactants, first order in H^+, independent of the concentration of benzoic acid, and free from salt effects (*25*). (The isotope content of the benzoic acid was determined by analysis of the carbon dioxide produced when the acid was decomposed over reduced copper at 500° in a nitrogen stream.)

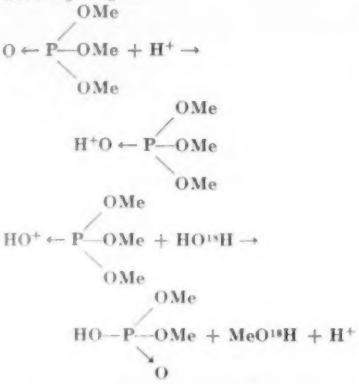
An interesting phenomenon was observed in the hydrolysis of trimethylorthophosphate (*26*) where the mechanism depended upon the pH of the solution. The two alternate mechanisms were represented as follows:

Alkaline Hydrolysis



$$OMe^- + HO^{18}H \rightarrow MeOH + O^{18}H^-$$

Acid Hydrolysis



A study of the biologically interesting anhydride, acetylphosphate, has shown that here again hydrolysis proceeds in two different ways, depending upon the pH of the solution (R. Bentley, in press). This is of interest since acetylphosphate has been widely considered as a possible acetylating and phosphorylating agent in physiological systems.

In these experiments, attention was concentrated on the analysis of the acetic acid produced, the dry silver salt being decarboxylated *in vacuo* and the carbon dioxide led into a mass spectrometer. It was of course necessary to take into account the exchange reaction of acetic acid itself, which proceeds in acid solution but not at all in alkaline solution. In alkaline solution, hydrolysis of acetyl phosphate took place by rupture of the carbon-oxygen bond (Case I) and, in acid solution, by rupture of the phosphorus-oxygen bond (Case II). See below.

The exchange reaction was greatly minimized in some experiments by the use of a very active acetylphosphatase preparation.* In slightly acid solution the hydrolysis was extremely rapid, and only a negligible amount of O^{18} was found in the acetic acid.

O^{18} has been extensively used by a number of Russian workers (Brodskiĭ, Dedussenko, Makolkin, Miklukhin) to investigate a number of organic reactions. These interesting results may be summarized as follows:

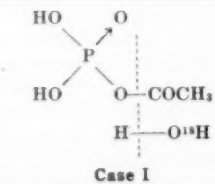
The reaction of acetic anhydride with ethyl alcohol takes place by the following mechanism:

$$CH_3CO\text{—}O\text{—}COCH_3 + H\,O^{18}C_2H_5 \rightarrow CH_3COO^{18}C_2H_5 + CH_3COOH$$

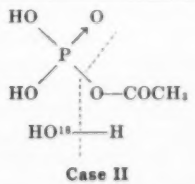
The heavy alcohol was obtained by fractionation using the previously described method of Urey; the analyses of ethyl acetate and alcohol were performed by mixing the sample with hydrogen and passing the mixture over thoriated nickel at high temperature.

A study of the Beckmann rearrangement of benzophenone oxime by phosphorus pentachloride in O^{18} enriched water showed that the reaction proceeded via a dehydration-hydration mechanism. The benzanilide formed was hydrogenated and shown to contain O^{18}; there was no exchange when benzanilide was treated with phosphorus pentachloride in enriched water. This provides additional proof for the theory that the Beckman rearrangement takes place by an ionic mechanism

* Supplied by Dr. F. Lipmann.

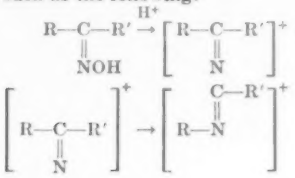


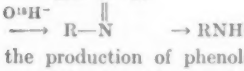
Case I



Case II

such as the following:





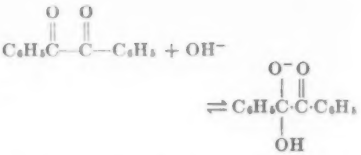
In the production of phenols from benzene sulphonic acid, β-naphthalene sulphonic acid, and β-anthraquinone monosulphonic acid by fusion with O^{18}-labeled sodium hydroxide, it was shown that the reaction took place via the addition of $O^{18}Na$ and elimination of SO_3Na. Water obtained from the phenols by hydrogenation contained excess isotope. Thus a rearrangement of the sulphonic salts to salts of ethers of sulphonic acids seems unlikely.

The xanthation of ethyl alcohol with carbon disulphide and $NaO^{18}H$ was shown to split the O—H bond of the alcohol.

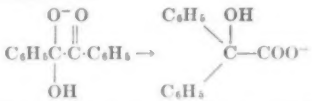
$$RO—H + HO^{18}Na + CS_2 \rightarrow RO{\cdot}CSSNa + H_2O^{18}$$

Water obtained from such sodium ethyl xanthate had the normal isotopic concentration. A similar reaction took place with alkali cellulose. Since the latter compound slowly exchanged its oxygen atoms with water, it was considered to be an addition product, $C_6H_{10}O_6{\cdot}NaOH$ (having the properties of a weak electrolyte) rather than an alcoholate, $C_6H_9O_4ONa$.

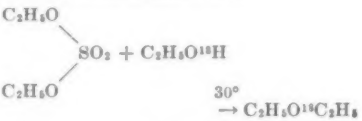
A study of the exchange of O^{18} between benzil and water provided information about the mechanisms of the hydroxyl ion catalyzed benzilic acid rearrangement (*27*). Benzil exchanged O^{18} much more rapidly in alkaline than in neutral solution and it was concluded that the first step in the rearrangement was the rapid reversible addition of OH^- to benzil.



This was followed by a slow rate-controlling rearrangement of the ion to benzilic acid.



The mechanism of ether formation from O^{18}-enriched ethyl alcohol has recently been studied (*28*).



After three and one-half hours, the O^{18}:O^{16} ratio in the ether was 85% that of the alcohol used. After thirty-five hours, however, this ratio had decreased to 60% that of the initial alcohol due to the reaction of more $(C_2H_5)_2SO_4$ with normal alcohol formed in side reactions.

Photosynthesis and O^{18} in the Atmosphere

An interesting observation was made by Dole in 1935. He showed that the oxygen of the atmosphere had a greater concentration of O^{18} than that of the oxygen derived from natural waters (*29*). This observation was subsequently confirmed by many other workers and is now known as the *Dole Effect*. It has also been demonstrated that the O^{18} content of natural carbonates is higher than in water.

In 1941, experiments were initiated on the study of photosynthesis using O^{18} as the tracer (*30*). Cell suspensions of *Chlorella pyrenoidosa* were allowed to photosynthesize in a buffer solution of bicarbonate-carbonate with a pH about 10. The over-all reaction is represented

by the usual equation:

$$CO_2 + H_2O + h\nu = O_2 + \frac{1}{n}(CH_2O)_n$$

When the photosynthetic process was carried out in water enriched with respect to the O^{18} isotope, the $O^{18}:O^{16}$ ratio of the evolved oxygen was equal to that of the water used. When the algae were grown in ordinary water containing enriched $KHCO_3^{18}$ and $K_2CO_3^{18}$, the isotope ratio of the evolved oxygen did not show any enrichment. (The rather slow exchange of O^{18} between carbon dioxide and water was, under the experimental conditions, sufficiently slow to be negligible by comparison to the rate of the photosynthetic process.) Similar results have also been obtained in the case of the land plants, sunflower and coleus.

These experiments show fairly conclusively that, in photosynthetic processes, the expired oxygen is derived from the water, and not from the carbon dioxide. Geochemical evidence is predominantly in favor of the view that atmospheric oxygen has been and is being produced by photosynthetic operations. Since about four fifths of the total photosynthesis takes place in the water of the oceans, it would therefore be expected that the O^{18} content of atmospheric oxygen would be equal to that of the water in the oceans, and different from that of naturally occurring carbon dioxide and carbonates. However, as was pointed out initially, it has been found that the reverse is actually true.

The first explanation of this phenomenon was that carbon dioxide, enriched in O^{18} by the well-known exchange reaction with water, contributed to the oxygen expired in photosynthesis. However, the experiments in which O^{18} was used to follow the course of photosynthesis show that this view is incorrect. Thus there is as yet no completely satisfactory explanation of the Dole Effect.

Two reactions which may be contributing to the enrichment are:

$O_2^{16}(g) + H_2O^{18}(g) = O_2^{18}(g) + H_2O^{16}(g)$
enrichment factor at 25° = 1.020

$O_2^{16}(g) + H_2O^{18}(l) = O_2^{18}(g) + H_2O^{16}(l)$
enrichment factor at 25° = 1.006

It is tempting to assume that the explanation may lie in the more favorable enrichment factor of the first equation, but near the earth's surface (where the oxygen was collected) the latter reaction is more likely to be the controlling factor. This is because of the large amount of liquid water compared to the amount of water vapor. The first reaction has been studied experimentally, and it was demonstrated that equilibrium was rapidly established on a hot wire at 1800° A, but more slowly at room temperature. The second reaction fails to account by itself, for the magnitude of the observed Dole Effect.

O^{18} in Metabolism Studies

With few exceptions, O^{18} has not been extensively used in studies of metabolic processes in physiological systems. One of its first applications in this field was a study of the fate of the sulphate radical in the animal body. It was employed in the form of heavy sodium sulphate (*14*). This was prepared from the reaction of sulphuryl chloride and H_2O^{18} using a catalytic quantity of iodine. The sodium sulphate was dissolved in normal water and injected into a rabbit; sulphate was recovered from the urine and analyzed for O^{18} as described earlier. The water prepared in this way showed a considerable concentration of O^{18}. After allowing for the ordinary sulphate excreted by the rabbit, it was concluded that most of the sulphate left the body unchanged. At most, only a small fraction of the ingested sulphate ions exchanged with other sulphate ions present beforehand.

It is to be expected that O^{18} will be more extensively used in the future

since it can be useful in the study of a variety of metabolic, organic and inorganic reactions whose intrinsic mechanisms are not completely understood.

BIBLIOGRAPHY

1. Giauque and Johnston, *J. Am. Chem. Soc.* **51,** 1436 (1929)
2. Babcock, *Proc. Natl. Acad. Sci.* **15,** 471 (1929)
3. Ruckärdt, *Naturwissenschaften* **18,** 534 (1930)
4. Murphey, *Phys. Rev.* **59,** 320 (1941)
5. Washburn, Smith, and Frandsen, *Bur. Standards J. Research,* **11,** 453 (1933); **13,** 599 (1934)
6. Selwood, Taylor, Hipple and Bleakney, *J. Am. Chem. Soc.* **57,** 642 (1935)
7. Stedman, *Can. J. Research* **13B,** 114 (1935)
8. Huffman and Urey, *Ind. Eng. Chem.* **29,** 531 (1937)
9. Urey, Mills, Roberts, Thode and Huffman, *J. Chem. Phys.* **7,** 138 (1939)
10. Thode, Smith and Walkling, *Can. J. Research* **22B,** 127 (1944)
11. Lewis, *J. Am. Chem. Soc.* **55,** 3502 (1933)
12. Clusius, Dickel and Becker, *Naturwissenschaften* **31,** 210 (1943)
13. Taylor and Bernstein, *J. Am. Chem. Soc.* **69,** 2076 (1947)
14. Aten and Hevesy, *Nature* **142,** 952 (1938)
15. Russell and Fulton, *Ind. Eng. Chem. Anal. Ed.* **5,** 384, (1933)
16. Datta, Day and Ingold, *J. Chem. Soc.,* 838 (1939)
17. Roberts and Urey, *J. Am. Chem. Soc.* **60,** 2391 (1939)
18. Herbert and Lauder, *Trans. Faraday Soc.* **34,** 432 (1938)
19. Polanyi and Szabo, *Trans. Faraday Soc.* **30,** 508 (1934)
20. Datta, Day and Ingold, *J. Chem. Soc.,* 1968 (1937)
21. Cohn and Urey, *J. Am. Chem. Soc.* **60,** 679 (1938)
22. Emeléus, James, King, Pearson, Purcell and Briscoe, *J. Chem. Soc.,* 1207 (1934)
23. Keston, Rittenberg and Schoenheimer, *J. Biol. Chem.* **122,** 227 (1937)
24. *Chem. Eng. News* **25,** 2515 (1947)
25. Roberts and Urey, *J. Am. Chem. Soc.* **61,** 2584 (1939)
26. Blumenthal and Herbert, *Trans. Faraday Soc.* **41,** 611 (1945)
27. Roberts and Urey, *J. Am. Chem. Soc.* **60,** 880 (1938)
28. Lauder and Green, *Nature* **157,** 767 (1946)
29. Dole, *J. Am. Chem. Soc.* **57,** 2731 (1935); *J. Chem. Phys.* **4,** 268 (1936)
30. Ruben, Randall, Kamen and Hyde, *J. Am. Chem. Soc.* **63,** 877 (1941)
31. Mears and Sobotka, *J. Am. Chem. Soc.* **61,** 880 (1939)

MANHATTAN DISTRICT DOCUMENTS

A NEW LISTING of Manhattan District Declassified Documents recently made public by the AEC appears below. This supplements the four lists previously published in NUCLEONICS.

Listed with the name of the paper are its author(s) and MDDC (Manhattan District Declassification Code) number. Papers which can be obtained from the Office of Technical Services, Department of Commerce, Washington, D. C., are noted by an asterisk (*). A reference appearing after an item indicates the journal in which the paper was published.

Crystal Structure Studies of Plutonium and Neptunium Compounds. W. H. Zachariasen. MDDC 67 (6/7/46)

Carcinogenic Action of Some Substances which May Be a Problem in Certain Future Industries. A. M. Brues, H. Lisco, M. Finkel. MDDC 145. *Radiology* **49,** 361 (1947)

Description of a Frequency Modulated Cyclotron and a Discussion of the Deflector Problem. E. J. Lofgren, B. Peters. MDDC 155 (7/2/46)

The Handling of Radioactive Materials in the Experimental Biology Section. P. C. Tompkins, A. Broido, J. D. Teresi. MDDC 377 (9/20/46)

Some Neutron Induced Activities in the Rare Earths. M. G. Inghram, R. J. Hayden. MDDC 428. *Phys. Rev.* **71,** 144 (1947)

Histopathology of Irradiation from External and Internal Sources. W. Bloom. MDDC 488. *Radiology* **49,** 344 (1947)

Effects of Total Surface Beta-irradiation. J. R. Raper, J. E. Wirth, K. K. Barnes. MDDC 499. *Radiology* **49,** 314 (1947)

The Mechanism of Disproportionation of

Pu(V). R. E. Conniek. MDDC 566 (12/23/46)

Components of the Acute Lethal Actions of Slow Neutrons. R. E. Zirkle. MDDC 585. *Radiology* **49,** 271 (1947)

Pilot Plant Synthesis of C_7F_{16}, C_8F_{16}, and Fluorocarbon Oils Boiling Up to 350° C. W. B. Burford, III. MDDC 600. *Ind. Eng. Chem.* **39,** 319 (1947)

The Clinical Sequences of Physiological Effects of Ionizing Radiation in Animals. C. L. Prosser. MDDC 611. *Radiology* **49,** 299 (1947)

Thermal Neutron Activation Cross Sections. L. Seren, H. N. Friedlander, S. H. Turkel. MDDC 613. *Phys. Rev.* **72,** 888 (1947)

Experience with Ionization Chamber Technique with C^{14} Activity. C. D. Janney, B. J. Moyer. MDDC 621

Geochemistry of Beryllium. M. Fleischer, E. N. Cameron. MDDC 643 (1/21/47)

Scattering of Protons by Deutrons. C. Bailey *et al.* MDDC 670. *Phys. Rev.* **72,** 662 (1947)

Spectral Emissivity and Total Emissivity of Beryllium Oxide. R. L. Seifert. MDDC 675 (2/19/47)

Certain Chemical and Physical Properties of the Uranium Chlorides. C. E. Evers. MDDC 690 (2/27/47)

The Effect of Adrenal Cortical Injury on the Toxicity of X-rays. B. N. Craver, M.D. MDDC 712 (12/31/36)

Metabolism of Plutonium. W. Langham *et al.* MDDC 719. *J. Biol. Chem.* **171,** 273 (1947)

Internal Cyclotron Targets. A. F. Reid. MDDC 730 (1/21/47)

Techniques in the Use of Carbon Fourteen. W. G. Dauben, J. C. Reid, P. E. Yankwich. MDDC 769 (3/18/47)

Method for the Preparation of Butt-Welded Thermocouples Using 3-Mil Diameter Wire. E. F. Hammel. MDDC 776 (4/3/47)

A Portable Pulse Generator. E. W. Titterton. MDDC 791. *Rev. Sci. Instr.* **18,** 639 (1947)

Short-lived Isomeric States of Se^{82} and Ge^{77}. J. R. Arnold, N. Sugarman. MDDC 837. *J. Chem. Phys.* **15,** 703 (1947)

Scattering of Neutrons in Polycrystals. R. J. Finkelstein. MDDC 845. *Phys. Rev.* **72,** 907 (1947)

The Structure of Uranium Hydride and Deuteride. R. E. Rundle. MDDC 865. *J. Am. Chem. Soc.* **69,** 1719 (1947)

Production and Properties of 50-year Element 94. G. T. Seaborg, A. C. Wahl, J. W. Kennedy. MDDC 867. *Phys. Rev.* **69,** 366 (1946)

The Diffraction of Neutrons by Crystalline Powders. E. O. Wollan, C. G. Shull. MDDC 869 (4/28/47)

The Critical Ionization Potentials of Uranium Hexafluoride and Hydrogen Fluoride. J. R. White, A. E. Cameron. MDDC 874. *Phys. Rev.* **71,** 907 (1947)

Biological Action of Gamma and X-rays. I. Exposure of Mice to Daily Doses of Gamma Radiation at two rates: 5.5 r/hr and 0.11 r/8 hr. E. Lorenz, A. Eschenbrenner, M. Derringer, W. E. Heston. MDDC 907. *J. Nat. Cancer Inst.* **6,** 349 (1946)

Economic Considerations. J. H. Lum. MDDC 909. *Chem. Eng.* **54,** 122 (1947)

Blood Platelet Counting. E. Skirmont. MDDC 916 (4/10/47)

Theory of the Synchrocyclotron. D. Bohm, L. L. Foldy. MDDC 933. *Phys. Rev.* **72,** 649 (1947)

Studies of the Delayed Neutrons. I. The Decay Curve and the Intensity of the Delayed Neutrons. A. H. Snell *et al.* MDDC 939. *Phys. Rev.* **7,** 541 (1947)

Studies of the Delayed Neutrons. II. The Chemical Isolation of the 56 Second and the 23 Second Activities. A. H. Snell *et al.* MDDC 940. *Phys. Rev.* **72,** 545 (1947)

Energy Distribution of the Fragments Resulting from the Fission of U^{235} and Th^{232} by Slow and by Fast Neutrons. J. L. Fowler, L. Rosen. MDDC 946. *Phys. Rev.* **72,** 926 (1947)

Angular Distribution of Neutrons from Targets Bombarded by 190 Mev Deuterons (BP-74). A. C. Helmholz, E. M. McMillan, D. Sewell. MDDC 951. *Phys. Rev.* **72,** 1003 (1947)

Cobalt 61 Radioactivity. T. J. Pharmley, B. J. Moyer. MDDC 957. *Phys. Rev.* **72,** 82 (1947)

Energy of Fission Fragments of U^{235}, U^{238}, Pu^{239}. T. Snyder. MDDC 959 (1947)

Effects of Radiation on Materials. A. O. Allen. MDDC 962 (5/20/47)

Distribution of C^{14} in Photosynthesizing Barley Seedlings. S. Aronoff, A. Benson, W. Z. Hassid, M. Calvin. MDDC 965. *Science* **105,** 664 (1947)

Neutron Cross Section Studies with the Rotating Shutter Mechanism. T. Brill, H. V. Lichtenberger. MDDC 968. *Phys. Rev.* **72,** 585 (1947)

A Neutron Detector Having Uniform Sensitivity from 10 Kev to 3 Mev. A. O. Hanson, J. L. McKibben. MDDC 972. *Phys. Rev.* **72,** 673 (1947)

2-inch Packless Through Vacuum Valve. R. Krohm. MDDC 974. *Rev. Sci. Instr.* **18,** 682 (1947)

The Halogen-Metal Interconversion Reaction and Its Application to the Synthesis

of Nicotinic Acid Labeled with Isotopic Carbon. A. Murray III, W. W. Foreman, W. Langham. MDDC 977 (5/13/47)

5.3 Day Element 61. G. W. Parker *et al.* MDDC 978. *Phys. Rev.* **72,** 85 (1947)

Adrenal Response to Total Body X-radiation. H. M. Patt, M. N. Swift, E. B. Tyree, E. S. John. MDDC 979. *Physiology* **150,** 480 (1947)

Frequency Modulated Cyclotron Characteristics. B. T. Wright, J. R. Richardson. MDDC 986*

Maintenance Experience and Development on the "California" Scaler. W. T. E. Elmendorf. MDDC 996 (5/14/47)

Summary of Metabolism of Carrier-Free Fission Products in the Rat. J. G. Hamilton MDDC 1002 (5/14/47)

Body Tissue Ionization due to Neutrons. H. A. Wilson. MDDC 1025 (5/29/47)

Note on Angular Distributions in Nuclear Reactions. E. Eisner, R. G. Sachs. MDDC 1029. *Phys. Rev.* **72,** 680 (1947)

A Laboratory Oscilloscope. V. Fitch, E. W. Titterton. MDDC 1033. *Rev. Sci. Instr.* **18,** 821 (1947)

Energy and Half Life of the Be^{10} *Radioactivity (BP-88).* E. M. McMillan. MDDC 1046. *Phys. Rev.* **72,** 591 (1947)

High Energy Photodisintegration of the Deuteron. M. E. Rose, G. Goertzel. MDDC 1049. *Phys. Rev.* **72,** 722 (1947)

The Clinical Sequence of Physiological Effects of Ionizing Radiation in Animals. C. L. Prosser *et al.* MDDC 1066. *Radiology* **49,** 299 (1947)

Artificial Radioactive Isotopes of Polonium. D. H. Templeton, J. J. Howland, I. Perlman. MDDC 1068. *Phys. Rev.* **72,** 758 (1947)

Thermal Neutron Flux in the Argonne Heavy-water Pile. W. H. Zinn. MDDC 1073 (6/24/47)

Vacuum Melting and Casting of Beryllium. E. Gordon. MDDC 1078. *Phys. Rev.* **72,** 387 (1947)

Cloud-chamber Energy Measurement of Photo-neutron Sources. D. J. Hughes, C. Eggler. MDDC 1082. *Phys. Rev.* **72,** 902 (1947)

The Isotopic Constitution of Lanthanum and Cerium. M. G. Inghram, R. J. Hayden, D. C. Hess, Jr. MDDC 1084. *Phys. Rev.* **72,** 967 (1947)

Mass Spectrographic Assignment of Activities to Re^{186} *and* Re^{188}. D. C. Hess, Jr., R. J. Hayden, M. G. Inghram. MDDC 1119. *Phys. Rev.* **72,** 730 (1947)

Formation and Properties of Radioactive Lithium. D. J. Hughes, D. Hall, C. Eggler, E. Goldfarb. MDDC 1120. *Phys. Rev.* **72,** 646 (1947)

Lethal Effects of X-rays on Four Marine Micro-plankton Organisms. K. Bonham *et al.* MDDC 1158. *Science* **106,** 245 (1947)

Decay Scheme of $_{45}Rh^{106}$. W. C. Peacock. MDDC 1169. *Phys. Rev.* **72,** 1049 (1947)

The 4n + 1 Radioactive Series: The Decay Products of U^{233}. F. Hagemann *et al.* MDDC 1186. *Phys. Rev.* **72,** 634 (1947)

Interaction of Neutrons with Electrons in Lead. W. W. Havens, Jr., I. I. Rabi, L. J. Rainwater. MDDC 1187. *Phys. Rev.* **72,** 634 (1947)

The Half-life of Tritium. M. Goldblatt, E. Robinson, R. W. Spence, MDDC 1129. *Phys. Rev.* **72,** 973 (1947)

Certain Aspects of the Acute Toxicity of Beryllium Following Intraperitoneal Injection. E. A. Maynard, W. L. Downs, H. C. Hodge. MDDC 1233 (7/2/47)

Half-life of Tritium. A. Novick. MDDC 1236. *Phys. Rev.* **72,** 972 (1947)

The Production of High Energy Neutrons by Stripping. R. Serber. MDDC 1238 (7/5/47)

The Neutron Scattering Cross Section at the Cadmium Resonance. W. W. Beeman. MDDC 1291. *Phys. Rev.* **72,** 986 (1947)

Resonance Scattering of Neutrons by Cobalt. S. P. Harris, A. S. Langsdorf, Jr., F. G. P. Seidl. MDDC 1299. *Phys. Rev.* **72,** 866 (1947)

Excitation Curve for the Reaction $C^{12}(p,pn)$ *Up to* 140 *Mev.* E. M. McMillan. MDDC 1306. *Phys. Rev.* **72,** 873 (1947)

Excitation Curves for the Reactions of $C^{12}(d,dn)C^{11}$ *and* $C^{12}(d,dn)C^{11}$ *at High Energies.* R. L. Thronton, R. W. Senseman. MDDC 1320. *Phys. Rev.* **72,** 872 (1947)

Effects of Space Charge on the Detection of High Energy Particles by Means of Silver Chloride Crystal Counters. L. F. Wouters, R. S. Christian. MDDC 1324. *Phys. Rev.* **72,** 1127 (1947)

Relative Cross Sections of Reactions Induced by High Energy Neutrons in C, N, O, and F. W. J. Knox. MDDC 1354 (10/3/47)

Nuclear Reactions at High Energies. R. Serber. MDDC 1357. *Phys. Rev.* **72,** 1114 (1947)

An Anomaly in the Study of Dielectric Constant. J. Van Wazer, I. Halpern. MDDC 1358 (10/14/47)

Isotope Techniques in Biochemistry — V

Most biochemical research problems employing isotope techniques require the synthesis of organic compounds from isotopic starting materials. This paper, the last of a series of five, discusses methods of synthesis useful for such work

By NORMAN S. RADIN

National Institute of Health Predoctorate Research Fellow
Biochemistry Department, Columbia University, New York

THE SYNTHESIS of isotopic compounds requires special consideration for several reasons:

(*a*) Isotopes are supplied as very simple compounds so that unfamiliar reactions must often be used.

(*b*) The expensiveness of isotopic starting compounds eliminates many otherwise suitable syntheses which give poor yields in terms of isotope content.

(*c*) Isotopes can be recovered from by-products by a combustion procedure, and a synthetic route should be chosen which gives a minimal dilution of the recovered isotope.

(*d*) Since supplies of isotopic compounds are limited, either because of expense or danger, reactions calling for a large excess of the isotopic compound cannot be used even though a good recovery can be obtained.

(*e*) It is usually desired to synthesize the compound with the label in only a certain position.

(*f*) The mechanism of some reactions is not entirely clear, so that it is sometimes necessary to check the location of the incorporated label.

(*g*) The frequent need for a maximal isotope concentration in the final product eliminates some otherwise suitable reactions which involve dilution of the isotopic starting material.

It is clear from these considerations, which will be illustrated later, that a rather new viewpoint must be adopted by the organic chemist who enters the isotope field.

The remainder of this paper is devoted to synthetic methods suitable for the more widely used isotopes.

HYDROGEN

Isotopic hydrogen can be obtained as deuterium oxide (practically 100% pure), deuterium gas, and tritium oxide. Although the last of these is not generally available, it may become so very soon. Both the heavy water and tritium oxide can be converted to gaseous hydrogen easily by electrolysis. In electrolyzing samples containing less than 100% isotope, appreciable enrichment of the heavier isotope occurs in the liquid phase, so that it is usually advisable to electrolyze the complete sample.

Methods for introducing labeled hydrogen into a compound can be divided into the following classes:

(1) Addition to a double or triple bond (with D_2, D_2O, DBr, etc.)

(2) Replacement of another element by deuterium

(3) Replacement of normal hydrogen atoms already present

(4) Biological synthesis.

Although most work has been done with

deuterium, it may be expected that the synthetic procedures apply to tritium about as well.

Addition Reactions

Reduction of an unsaturated bond is a convenient and efficient method of introducing tagged hydrogen into a desired position. The reduction can be performed catalytically with molecular deuterium or chemically in the presence of heavy water. Although no suitable micro-method for high-pressure reduction has been developed, this is not a serious handicap. Modern catalysts are remarkably active and make possible a large variety of reductions at pressures near one atmosphere. Since the methods are well known and obviously of wide usefulness, no descriptions will be given here. An incomplete summary of syntheses using isotopic hydrogen may be found in Kamen's monograph (*1*).

The unsaturated bond that is reduced need not be carbon-carbon but can also be carbon-oxygen or carbon-nitrogen. For example, labeled glutamic acid can be made by reductive amination of α-ketoglutaric acid (*2, 3*). The acid is shaken with ammonia and palladium in the presence of isotopic hydrogen, with the introduction of the labeled atoms into the α- *and* β-positions. The unexpected appearance of isotope at the β-carbon points up the need for locating the positions of the isotopic atoms, particularly if the reaction mechanism is not entirely certain. Amino acids can also be made by reduction of the oximes and phenylhydrazones of α-keto acids (*4*).

If possible, aqueous or alcoholic media should be avoided, inasmuch as some samples of catalyst are able to catalyze the exchange between gaseous hydrogen and the ionizable hydrogen atoms of the solvent. This results in a reduction of the isotope concentration of the gaseous hydrogen.

Lithium aluminum hydride (*5*), the new reducing agent, would be very useful if made with isotopic hydrogen. This compound reduces esters, acid chlorides and ketones to alcohols, and nitriles to amines, without affecting any carbon-carbon double bonds that might be present. Other reducing agents, such as aluminum amalgam, sodium amalgam and chromous chlorid can be used with heavy water as the source of deuterium.

A compound may be required in which the isotopic hydrogen is attached to only one carbon atom. Reduction of an unsaturated C—C linkage can be used if the hydrogen on one of the carbons is labile under appropriate conditions. In this case, the unwanted isotopic atoms can be "washed out" by exchange with a large amount of normal hydrogen. For example, one could probably prepare 1-deuterio-3-butanone by partial reduction of vinylmethylketone and subsequent removal of the deuterium on carbon-2 by treatment with alkali in a large volume of water. The conditions under which labilization occurs are discussed in the third paper in this series (*6*).

Another method of introducing only one isotopic hydrogen atom is the addition of DBr to a double bond. The Markownikoff rule can often be reversed, if desired, by the addition of some organic peroxide. Dry hydrogen bromide can be made by heating deuterium gas with bromine (*7*), or by adding heavy water to thionyl bromide (*8*) or phosphorus tribromide. Deuterium chloride is easily made by adding heavy water to thionyl chloride (*9, 10*).

It is also possible sometimes to add water directly to an unsaturated bond. An outstanding example is the hydration of acetylene to acetaldehyde (*11,12*).

Replacement of Another Element

There are a number of simple syntheses in which isotopic hydrogen re-

places an atom of another element. For instance, decarboxylation reactions under anhydrous conditions result in movement of the carboxyl hydrogen to the α-carbon. Carboxyl-labeled compounds are easily prepared by hydrolysis of acid anhydrides or chlorides in heavy water. Acetaldehyde, labeled only in the CHO group, can be made by decarboxylating pyruvic acid in heavy water. Deuterio benzene has been prepared by decarboxylation of calcium mellitate in presence of $Ca(OD)_2$ (*14*).

Replacement of halogen atoms is frequently simple, by reduction with isotopic hydrogen and palladium black (or Pd-$BaSO_4$), or by formation of the Grignard reagent and reaction with labeled water.

A useful reaction is the replacement of calcium in calcium carbide to form acetylene (*12, 15*). Aromatic sulfonic acids can be converted to the hydrogen derivatives by heating with water.

Hydrogen Exchange Reactions

One of the most useful of labeling techniques is that peculiar to the isotope chemist, the exchange of protium by deuterium or tritium. Any compound containing an activated carbon atom can be treated with an appropriate isotopic compound and exchange effected. The simplest example is found in the alkali-catalyzed exchange between heavy water and the enolizing hydrogens of ketones (*16*). Aromatic compounds will exchange ring hydrogens when treated with aluminum chloride and DCl (*10*).

An elegant synthesis of methyl-labeled acetic acid involves exchange of the methylenic hydrogens of malonic acid in heavy water and subsequent partial decarboxylation to acetic acid (*17*). This synthesis also illustrates the technique of subsequent stabilization of the introduced deuterium atoms. The isotopic atoms introduced by exchange might exchange during the course of the biological experiment, with consequent loss of the labels, so that it is advisable to remove or modify the labilizing group.

It has been found that exchange will take place with substances ordinarily considered inactive by heating with concentrated deuterium sulfate (*58*). This method has been used for aromatic compounds, in which the ring is preferentially substituted (*18*), with several amino acids (*19*) and with fatty acids (*20*). In the case of the fatty acids, the isotopic hydrogen is introduced only on the α-carbon. Deuterium sulfate is readily made in small amounts by hydration of SO_3. This may be carried out in two beakers, one containing SO_3, the other D_2O, in an evacuated desiccator.

Heating with heavy water and a suitable catalyst will result in considerable exchange for a number of substances. Fatty acids, treated at 130° with alkaline deuterium oxide and platinum for a week, take up much more deuterium than by the above-mentioned method (*20*). The deuterium atoms seem to be distributed evenly throughout the carbon chain.

Cholesterol can be labeled by heating with heavy water, together with acetic acid and active platinum (*21*). In this case, the side chain exchanges more than the ring structure. The total empiricism of the technique is shown by the inactivity of cholesteryl chloride under the same conditions, while desoxycholic acid exchanges readily even without the addition of the acetic acid. Selenium, a dehydrogenator, produced a little exchange in cholesterol. A certain amount of danger exists in the application of this method to unsaturated compounds, since platinum tends to catalyze the movement of double bonds.

Exchange methods of labeling obviously have the drawback that dilution of the source isotope must take

place. Since deuterium is now supplied in almost 100% concentration, this dilution is often not important. The low price of deuterium permits use of a large excess so that dilution is minimized. Tritium oxide will probably be generally available in very high concentrations also.

Another drawback is that labeling of specific carbon atoms cannot always be managed, nor are the positions actually labeled always known. If necessary, localization studies can be made. The main point in favor of hydrogen exchange is the ease with which complex isotopic molecules can be prepared. Some substances that at present cannot be synthesized at all, such as cholesterol, can be labeled readily.

Biological Synthesis

By maintaining an organism in a *milieu* containing isotopic water, labeled compounds can be obtained simply by isolation. This technique resembles the exchange techniques in its advantages and disadvantages. The positions of the tagging atoms and their concentrations depend on the organism and nature of the diet. Fatty acids isolated from rats fed heavy water will have a higher isotope concentration when a high carbohydrate diet rather than a high fat diet is used; the reverse would probably be found in the case of liver glycogen.

Biological syntheses tend to be wasteful, since the few compounds that may be isolated from the organism usually contain only a small part of the isotopic water expended. Of course, the isotope may be recovered, more or less diluted, by combustion of the remaining material. It is sometimes possible to choose organisms, particularly in the lower groups, that will give especially high yields (*22*).

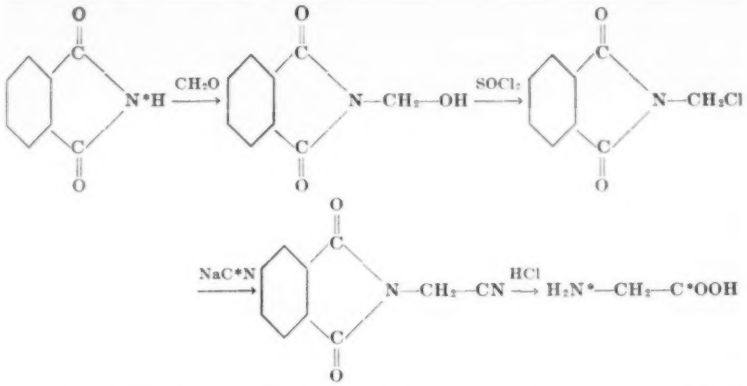
With the higher organisms, specific positions can sometimes be labeled by feeding an isotopic compound known to be a precursor of the desired substance. For example, glutathione might be made labeled only in the cysteine part by feeding isotopic cysteine and isolating the tripeptide. A very small amount of isotopic hydrogen might be found in the glycine and glutamic acid parts. This technique is more useful for labeling with other elements, since hydrogen tends to be lost more quickly from the carbon atoms. The use of a *Neurospora* mutant requiring a special compound permits labeling conversion products of that compound with essentially no dilution (*23*).

A special type of biological synthesis is the use of enzyme preparations or tissue slices. Fumaric acid, labeled with C^{11}, was prepared quickly by allowing a succinic dehydrogenase preparation to act on isotopic succinic acid (*24*). A variation of this technique can be applied to reversible enzymatic reactions: Allow an enzyme or tissue slice to act on the desired, *non*isotopic substance in the presence of an isotopic breakdown product. On stopping the reaction before reaching equilibrium and isolating the original substrate, it will be found that the desired compound contains a certain amount of isotope. The actual concentration will depend on the ratio of the rates of the forward and reverse reactions.

The dilutions often obtained in biological and exchange methods can be made less of a handicap by the use of tritium instead of deuterium because the former can be diluted very much more without loss of precision of measurement. This is true in general of all radioactive as compared with stable isotopes of the same element.

NITROGEN

Heavy nitrogen can be obtained in concentrations as high as 60% N^{15} in the form of ammonium salts and potassium phthalimide. Since this is



one of the most expensive isotopes, it is necessary to emphasize good yields, efficient recovery without much dilution, and low initial investments. The last requirement eliminates many syntheses that call for a large excess of ammonia.

Most work with N^{15} has involved the synthesis of amino acids. Here the Knoop-Oesterlin reduction in the presence of ammonia and the replacement of α-bromine atoms by the phthalimide group have been quite effective (*3*). The reduction method uses α-keto acids, which are shaken with two equivalents of heavy ammonia (three in the case of the dicarboxylic acids), palladium, and hydrogen in a mixture of alcohol and water. The only side reaction seems to be reduction of the keto acid to the hydroxy acid; this is probably minimized by using a low temperature and a suitable catalyst.

The Gabriel phthalimide synthesis may be conducted with the bromo or chloro esters by heating to 150–200° with well-powdered potassium phthalimide, sometimes in the presence of some cupric oxide. The reaction is exothermic, so that large runs must be made with care. The α-phthalimido ester that forms is hydrolyzed completely by refluxing with a mixture of acetic and hydrochloric acids.

The phthalimide NH group is rather reactive and will add on to activated double bonds as well as the carbonyl bond. The latter reaction in the preparation of doubly labeled glycine (*25*) is illustrated above.

Potassium phthalimide will react with bromomalonic ester to yield the useful phthalimidomalonic ester, an intermediate which will react in the presence of sodium ethylate with organic halides to produce, eventually, α-amino acids. N^{15}-proline has been prepared in this way, using trimethylene bromide as the halide. Hydrolysis of the condensation product yields a bromo amino pentanoic acid which cyclizes to form proline (*26*).

A method of amination that may prove useful for certain cases is that based on the reaction of benzylamine and halides. The benzyl group serves to protect the amine if other reactions are to follow; at the proper time the benzyl radical is removed by hydrogenolysis rather than hydrolysis, as in the use of phthalimide. This method has been used in a recent synthesis of serine (*27*).

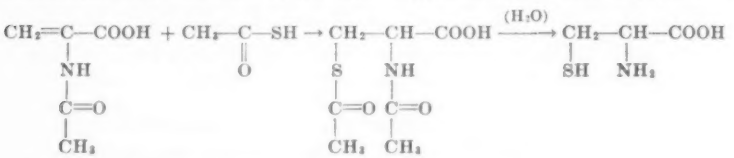
The use of the Gabriel phthalimide

reaction for tertiary halides tends to produce unsaturated compounds; with aromatic halides poor yields are obtained. In these cases it might be possible to adapt the Coleman-Hauser reaction of Grignard reagents with chloroamine to isotopic work (*28*):

$$NaOCl + NH_3 \rightarrow Cl{-}NH_2$$
$$Cl{-}NH_2 + R{-}Mg{-}Cl \rightarrow R{-}NH_2 + MgCl_2$$

In general, aliphatic and aromatic amines are probably best prepared by a Hofmann hypobromite reaction with an amide. The amide is easily prepared by reaction of heavy ammonia with an acid chloride or anhydride. Anthranilic acid has been synthesized in the Columbia University laboratory from isotopic potassium phthalimide in this way (*29*).

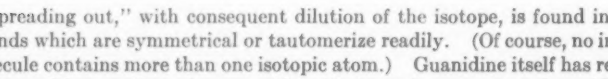
An unusual amination reaction which may prove useful for the synthesis of cysteine (*49, 50*) is the condensation of acetamide with pyruvic acid to yield α,α-diacetamidopropionic acid. This can be broken down partially to α-acetamidoacrylic acid, which can be converted to cysteine by the following reactions:



A useful intermediate, cyanamide, has been prepared from heavy ammonia (*30*). Dry methanolic ammonia is allowed to react at room temperature with a solution of cyanogen bromide in ether:

$$2NH_3 + Br{-}CN \rightarrow NH_2{-}CN + NH_4Br$$

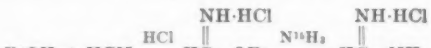
Isotopic carbon could be introduced here, perhaps, by allowing bromine to react with aqueous sodium cyanide and subsequently distilling out the cyanogen bromide. Although only the amine nitrogen in the cyanamide is labeled, condensation with sarcosine gives creatine in which the guanidine group is labeled in *both* nitrogens (*30, 31*):



This "spreading out," with consequent dilution of the isotope, is found in many compounds which are symmetrical or tautomerize readily. (Of course, no individual molecule contains more than one isotopic atom.) Guanidine itself has recently been prepared by reaction of cyanamide with isotopic ammonium bromide (*13*). Here, all three nitrogen atoms are labeled.

Cyanamide can also be converted to O-methylisourea, a useful intermediate, by treatment with methanol and hydrogen chloride (*32, 46*). N^{15}-containing O-methylisourea has been used to prepare arginine labeled in the guanidine nitrogens by reaction with α-tosyl ornithin (*33*).

Isotopic formamidine has been prepared by the usual reactions (*13*):

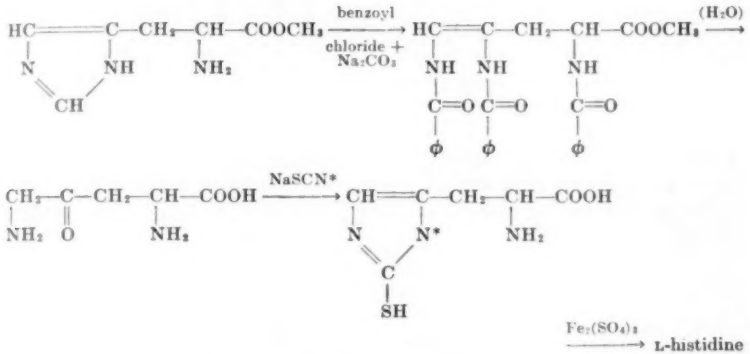


The formamidine was used in the synthesis of adenine. Cyanamide, guanidine, O-methylisourea and formamidine find use in the preparation of heterocyclic compounds.

Isotopic sodium thiocyanate is a useful reagent that can be made from heavy ammonia in good yield (*34*). The reaction proceeds mainly according to the equation:

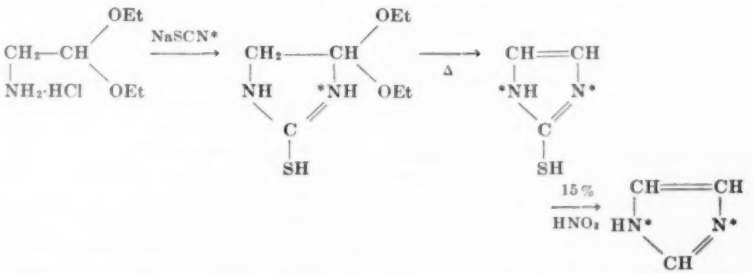
$$3NH_4NO_3 + 6NaOH + 3CS_2 + 2Fe(OH)_3 \longrightarrow 3NaSCN + 2FeS + S + 3NaNO_3 + 12H_2O$$

This thiocyanate has been used for the synthesis of L-histidine starting with natural, optically active histidine:



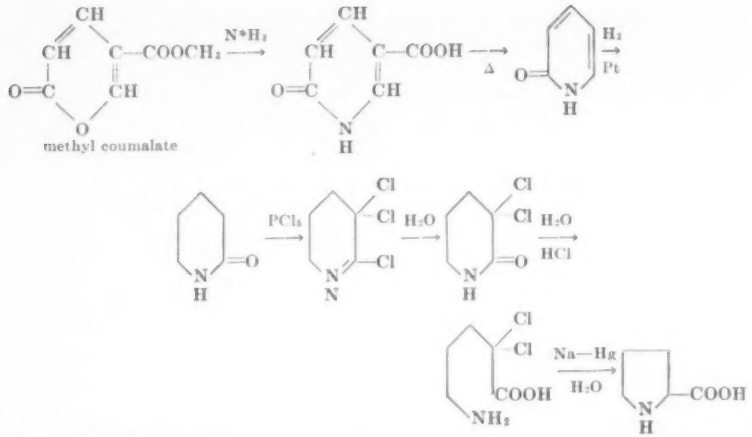
This synthesis is especially interesting because it illustrates the technique of partial breakdown, especially useful for compounds that are difficult to synthesize but relatively easy to isolate from natural sources. Another excellent example of this technique is found in the synthesis of cholestenone with one carbon in the A-ring labeled (*35*).

The parent compound, imidazole, has been prepared from N^{15}-thiocyanate by a similar condensation reaction (*36*):



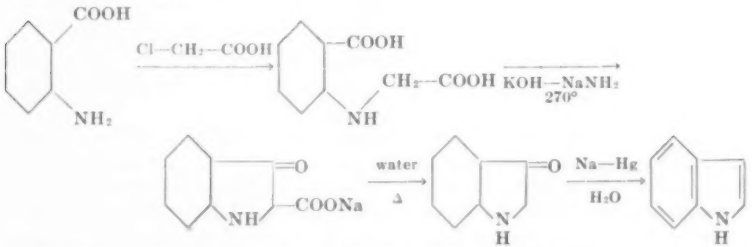
Note the "spreading out" of the label when cyclization occurs.

Hydroxynicotinic acid, other pyridine derivatives, and proline have been prepared with isotopic nitrogen by an interesting method (*37*):



It seems likely that the replacement of oxygen in heterocyclic compounds by ammonia can serve to produce a wide variety of nitrogen-ring compounds.

Another heterocyclic compound, indole, has been prepared from isotopic anthranilic acid (*29*):



The indole can be used for the synthesis of ring-N^{15} tryptophane.

While all the above reactions are run at atmospheric pressure, there are many reactions involving ammonia (as well as compounds of other elements) which would be useful if micro high-pressure reactions could be conducted readily. A micro hydrogenation bomb can be used for many preparations, the high pressure being attained by filling the bomb with liquid ammonia and then heating it. It is often more convenient to use a glass bomb tube, particularly when it is desired to fill the tube while connected to a high-vacuum manifold. When pressures too great for the glass are needed, the sealed tube can be placed in a metal hydrogenation bomb which is then filled with an inert gas to approximately the pressure expected inside the reaction tube. This technique has been used in a recent synthesis of C^{14}-urea (*38*).

Exchange reactions like those found so useful for deuterium do not seem to have been studied for nitrogen. The only experiments known to the author are those conducted under mild conditions preliminary to the first use of N^{15} as a tracer (*39*). It might prove interesting to attempt exchanges between

ammonia and aminoid compounds, including heterocyclic compounds, at high temperatures and pressures.

Biological syntheses have also been relatively neglected in the case of N^{15}, although several advantages over isotopic hydrogen and carbon exist. When microorganisms are used, the waste nitrogen can be recovered easily and often without dilution by the Kjeldahl procedure. Many organisms will grow on ammonia as the sole source of nitrogen, so that there is no ambiguity as to the position of the isotope. Moreover, many nitrogen compounds have only one nitrogen atom per molecule, so that the common objection to hydrogen and carbon isotopic biosyntheses—indiscriminate labeling—is avoided.

There is probably a demand for the oxidized forms of heavy nitrogen, perhaps more for organic than biochemical research. Efficient syntheses of hydroxylamine and nitrite esters from ammonia would probably find considerable use. Aromatic amines can be converted to nitro compounds by oxidation, using persulfuric acid, hydrogen peroxide, or benzoyl peroxide.

CARBON and OXYGEN

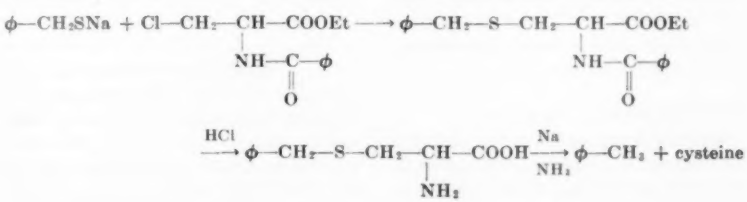
The synthesis of compounds with isotopic carbon has been discussed in detail in a recent review in NUCLEONICS by Miller and Price (*40*). Heavy oxygen syntheses are considered in a review by Ronald Bentley elsewhere in this issue; see page 18.

SULFUR

Radioactive sulfur can be obtained from the U. S. Atomic Energy Commission in the forms of sulfuric acid and sodium sulfide. Stable sulfur isotopes are not generally available for purchase, but can become so if the demand is sufficient. Of the two forms, sodium sulfide is the more generally useful, since almost every type of organic sulfur compound can be made from it, more or less directly.

Sodium sulfide, or rather sodium hydrosulfide, will react readily with active halogen compounds to form mercaptans. The most useful of these is benzyl mercaptan, which, in analogy to benzylamine, reacts readily with halides to produce benzyl-thio ethers. From these the benzyl group can be removed by treatment with sodium in liquid ammonia. In this way, homocysteine and cysteine have been prepared (*41*, *42*, *43*, *48*). In a typical reaction, shown below, the isotopic sulfur is incorporated quite near the end of the synthesis to reduce losses.

An alternate synthesis for benzyl mercaptan is by the reaction of benzyl magnesium chloride with free sulfur (*42*). The sulfur is made from sodium sulfide in almost quantitative yield by oxidation with a mixture of iodine, potassium iodide, and hydrochloric acid. The Grignard reaction is probably the best method for the preparation of aromatic mercaptans, which can then be converted to sulfonic acids and other derivatives.



A recent study has been reported in which the biological fate of the thiocyanate ion was traced (*44*). The S^{35}-containing thiocyanate was synthesized by heating free sulfur with potassium cyanide in acetone. This thiocyanate might be useful for the synthesis of isotopic ergothioneine by a variation of the Tesar-Rittenberg synthesis of N^{15}-histidine (*34*).

Radioactive sulfate has been used for a biological synthesis of glutathione (*45*). A yeast known to form much glutathione was grown on a medium containing half its sulfur in the form of $MgS^{35}O_4$. From 30 gm of yeast, 20 mg of the cuprous salt of the tripeptide was isolated.

In this connection, it might be pointed out that very high radioactivity intensities can be used in biological syntheses as long as the damage that ensues in the cells does not interfere with the synthesis. The isolation procedure can then be eased considerably by adding nonisotopic carrier; the moderate dilution that thus occurs often does not depreciate the usefulness of the compound.

It should be remembered that the half-life of S^{35} is only 87 days, so that very long-term syntheses and biological experiments must be performed with a stable isotope.

In general, it is possible to recover sulfur wastes by fusion with sodium peroxide (*43*). The sodium sulfate can then be purified through conversion to the benzidine salt and the sulfate either reduced to sulfide by heating with formic acid, hydriodic acid and red phosphorus (*42*), or by heating barium sulfate in a stream of hydrogen at 800–1000° (*47, 48*).

PHOSPHORUS

Radioactive phosphorus is sold in the form of sodium phosphate by the Atomic Energy Commission. The great majority of biochemical experiments have used sodium phosphate directly, in part because the omnipresent phosphatases rapidly hydrolyze any synthetic phosphate esters or anhydrides that might be administered. Isotopic organic phosphates may prove useful in certain types of experiments, however, and a brief description of published synthetic methods will be given.

Radioactive phosphoric acid has been used for the synthesis of phosphoryl choline, the acid being mixed with choline chloride and maintained at 165° *in vacuo* over P_2O_5 (*52*). A more generally useful phosphorylating agent is phosphoryl chloride, which can be prepared by heating isotopic silver phosphate with normal phosphorus pentachloride in a bomb tube at 130° for ten minutes (*53*). The $POCl_3$ that is formed is diluted, of course, but the high specific activity of the available phosphate makes this unimportant.

Radioactive phosphoryl chloride has been used for the synthesis of α-glycerophosphoric acid, the esterification being performed on acetonyl glycerol which is then partially hydrolyzed to remove the protecting group (*54*). By adding aqueous ethanolamine to phosphoryl chloride, ethanolamine phosphate can be prepared (*53, 56*). An efficient synthesis of radioactive diphenyl phosphoryl chloride would probably be quite useful.

Considerable work with P^{32}-phosphate metabolism has resulted in much literature which should be consulted with biological synthesis in mind. The reader is recommended to the experiments of Parnas (*55*) and the exhaustive review by Hevesy (*57*).

General Considerations

Double labeling—the use of two different isotopes in the same compound—has many advantages, and increasing use of the technique can be expected. Not only is information about the two different atoms obtained,

but also a measure of their relative reaction rates. The use of controls in quantitative metabolic studies comparing the fates of the two labeled atoms is made unnecessary. So much effort usually goes into the biological and isolation parts of a problem that a maximum of information should be obtained per experiment. It is often possible to obtain greater yields of the desired compound by synthesizing it in two different ways, incorporating only one isotope in each path. Mixing the two products gives the doubly labeled molecule, albeit with both isotopes diluted. If sufficiently large batches are made, samples of both products can be saved for experiments requiring only one isotope.

In some experiments, particularly metabolic conversion studies, the presence of only a trace of an isotopic impurity in the synthetic product may yield misleading results. The reader is reminded of the washing-out technique and the various purity criteria described in previous papers in this series. An interesting example of the washing-out technique is found in a resolution of DL-leucine, containing N^{15} (*51*). The crude brucine salt of isotopic formyl-L-leucine, obtained by fractional crystallization, was recrystallized in the presence of the brucine salt of *non*isotopic formyl-D-leucine. Although the final product still contained some contaminating D-isomer, the isotope concentration in the impurity had been lowered considerably by the admixture of the nonisotopic material. This method, which has wide applicability, cannot be used if a solid solution is formed by the desired substance and its contaminant.

A simple method of increasing the yield of labeled material consists of adding unlabeled material to the mother liquors and recrystallizing. Although the extra crop that is obtained has a lower isotope concentration than the main crop, it may well find subsequent experimental use.

When a racemic mixture is produced at the end of an isotopic synthesis and only one optical isomer is wanted, it is frequently convenient to add a nonisotopic optically active sample of the same substance and isolate the added material, which is now labeled. For example, rather than resolve labeled DL-glutamic acid, one can simply add some normal L-glutamic acid (isolated from natural sources) and, using the proper volume of 6*N* HCl, crystallize out L-glutamic acid hydrochloride containing some isotope. (In the case of glutamic acid, the yield of L-isomer is occasionally more than was added.) Of course this method is wasteful of isotopic starting material and gives a lower isotope concentration than could be obtained by resolution, but some resolutions are quite tedious or give low yields, and the cost of the isotope may be offset by the saving in time. Syntheses involving radioactive isotopes usually call for a deliberate dilution at some point, anyway.

In some cases, the presence of the other optical isomer is not objectionable as long as it is not labeled. It is possible then simply to discard the unwanted labeled isomer by several recrystallizations of the racemic mixture in the presence of large amounts of the unwanted *un*labeled isomer. For example, tagged L-glutamic acid molecules can be washed out of the DL-amino acid by adding normal L-glutamic acid and crystallizing out the L-acid hydrochloride. After several dilutions, the remaining DL-glutamic acid hydrochloride will have the isotope in only the unnatural form.

Laboratory Techniques

A technique in isotopic synthesis that is coming into greater favor is the use of a high-vacuum manifold. The new C^{14} literature contains many descrip-

tions of this technique. In general, it involves the use of a long, wide tube, connected at one end to a mercury-diffusion pump, with several side tubes to which reaction and storage vessels and manometers are attached by standard taper joints. Gases are transferred by displacing with mercury or cooling the receiving vessel with liquid nitrogen. Liquids are transferred by vacuum distillation or sublimation after freezing. The advantages of this technique are that very small quantities can be handled quickly and without loss, dangerous radioactive isotopes are kept enclosed until the end of the reaction, and unstable compounds are safely distilled at the low pressures used.

The enormous range of radioactivity-measuring devices makes the danger of contamination quite serious for the radioisotopes. For this reason, it is advisable to conduct syntheses with the radioelements in two separate laboratories, one for high-activity samples, the other for low. Isolations of biological materials should be conducted in still another laboratory. Other steps for the minimizing of contamination consist in using separate apparatus and reagents in the different laboratories, glass or stainless steel bench tops, efficient hoods, and portable radiation detectors (monitors). Disregard for such precautions in a laboratory using long-lived radioisotopes may result in such wide-spread contamination that either moving, or grimly thorough housecleaning, will be necessary.

BIBLIOGRAPHY

1. M. D. Kamen, "Radioactive Tracers in Biology," Chapt. 6, (Academic Press, Inc., New York, 1947)
2. D. Rittenberg, S. Ratner, H. D. Hoberman, *J. Am. Chem. Soc.* **62,** 2249 (1940)
3. R. Schoenheimer, S. Ratner, *J. Biol. Chem.* **127,** 301 (1939)
4. V. V. Feofilaktov, V. N. Zaitseva, *J. Gen. Chem.* (*U.S.S.R.*) **10,** 1391 (1940); E. Fischer, R. Groh, *Ann.* **383,** 363 (1911); K. E. Hamlin, Jr., W. H. Hartung, *J. Biol. Chem.* **145,** 349 (1942); D. Shemin, R. M. Herbst, *J. Am. Chem. Soc.* **60,** 1951 (1938)
5. R. F. Nystrom, W. G. Brown, *J. Am. Chem. Soc.* **69,** 1197 (1947); A. E. Finholt, A. C. Bond, Jr., H. I. Schlesinger, *J. Am. Chem. Soc.* **69,** 1199 (1947)
6. N. S. Radin, NUCLEONICS, Vol. **1,** No. 4, 51 (1947)
7. C. L. Wilson, A. W. Wylie, *J. Chem. Soc.* 596 (1941)
8. R. C. Elderfield, W. J. Gensler, F. Brody, J. D. Head, S. C. Dickerman, L. Wiederhold III, C. B. Kremer, H. A. Hageman, F. J. Kreysa, J. M. Griffing, S. M. Kupchan, B. Newman, J. T. Maynard, *J. Am. Chem. Soc.* **68,** 1579 (1946)
9. T. P. Nevell, E. de Salas, C. L. Wilson, *J. Chem. Soc.* 1188 (1939)
10. A. Klit, A. Langseth, *Z. physik. Chem.* **176A,** 65 (1936)
11. K. Bloch, D. Rittenberg, *J. Biol. Chem.* **155,** 243 (1944)
12. J. E. Zanetti, D. V. Sickman, *J. Am. Chem. Soc.* **58,** 2034 (1936)
13. G. B. Brown, P. M. Roll, A. A. Plentl, *Federation Proc.* **6,** 517 (1947)
14. H. Erlenmeyer, H. Lobeck, *Helv. Chim. Acta* **18,** 1464 (1935)
15. W. I. Patterson, V. du Vigneaud, *J. Biol. Chem.* **123,** 327 (1938)
16. M. Anchel, R. Schoenheimer, *J. Biol. Chem.* **125,** 23 (1938)
17. J. O. Halford, C. L. Anderson, *J. Am. Chem. Soc.* **58,** 736 (1936)
18. A. R. Moss, R. Schoenheimer, *J. Biol. Chem.* **135,** 415 (1940)
19. D. Rittenberg, A. S. Keston, R. Schoenheimer, G. L. Foster, *J. Biol. Chem.* **125,** 1 (1938)
20. W. E. van Heyningen, D. Rittenberg, *J. Biol. Chem.* **125,** 495 (1938)
21. K. Bloch, D. Rittenberg, *J. Biol. Chem.* **149,** 505 (1943)
22. J. L. Stokes, M. Gunness, *J. Bact.* **52,** 195 (1946)
23. G. B. Brown, Report of a Symposium on the Use of Isotopes in Biological Research, March 1947, p. 40; Committee on Growth, National Research Council
24. M. B. Allen, S. Ruben, *J. Am. Chem. Soc.* **64,** 948 (1942)
25. W. Sakami, W. E. Evans, S. Gurin, *J. Am. Chem. Soc.* **69,** 1110 (1947); D. Shemin, *J. Biol. Chem.* **162,** 297 (1946)
26. D. Shemin, *J. Biol. Chem.* **162,** 297 (1946)
27. A. M. Mattocks, W. H. Hartung, *J. Biol. Chem.* **165,** 501 (1946)
28. C. Weygand, "Organic Preparations," p. 214 (Interscience Publishers, Inc., New York, 1945)
29. R. W. Schayer, unpublished work
30. K. Bloch, R. Shoenheimer, D. Rittenberg, *J. Biol. Chem.* **138,** 155 (1941)
31. K. Bloch, R. Schoenheimer, *J. Biol. Chem.* **131,** 111 (1939)
32. J. Steiglits, R. H. McKee, *Ber. chem. Ges.* **33,** 1517 (1900)
33. K. Bloch, R. Schoenheimer, *J. Biol. Chem.* **138,** 167 (1941)
34. C. Tesar, D. Rittenberg, *J. Biol. Chem.* **170** 35 (1947)
35. R. B. Turner, *J. Am. Chem. Soc.* **69,** 726 (1947)

36. N. S. Radin, D. Shemin, unpublished work.
37. M. R. Stetten, R. Schoenheimer, *J. Biol. Chem.* **153,** 113 (1944)
38. H. E. Skipper, C. E. Bryan, O. S. Hutchinson, Atomic Energy Commission Isotopes Division Circular C-8 (1947)
39. A. S. Keston, D. Rittenberg, R. Schoenheimer, *J. Biol. Chem.* **127,** 315 (1939)
40. W. W. Miller, T. D. Price, NUCLEONICS Vol. **1**, No. 3, 4 (1947); Vol. **1**, No. 4, 11 (1947)
41. J. B. Melchior, H. Tarver, *Arch. Biochem.* **12,** 301 (1947); W. I. Patterson, V. du Vigneaud, *J. Biol. Chem.* **111,** 393 (1935)
42. G. W. Kilmer, V. du Vigneaud, *J. Biol. Chem.* **154,** 247 (1944)
43. H. Tarver, C. L. A. Schmidt, *J. Biol. Chem.* **130,** 67 (1939)
44. J. L. Wood, E. F. Williams, Jr., N. Kingsland, *J. Biol. Chem.* **170,** 251 (1947)
45. R. G. Franklin, *Science* **89,** 298 (1939)
46. J. Kapfhammer, H. Mueller, *Z. physiol. Chem.* **225,** 1 (1934)
47. F. C. Henriques, Jr., C. Margnetti, *Ind. Eng. Chem., Anal. Ed.* **18,** 476 (1946)
48. H. Tarver, C. L. A. Schmidt, *J. Biol. Chem.* **146,** 69 (1942)
49. M. W. Farlow, U. S. Patent 2,406,362 (1946); *Chem. Abstracts* **40,** 7233 (1946)
50. D. Shemin, Isotopes Symposium (see ref. 23), p. 31; M. Bergmann, K. Grafe, *Z. physiol. Chem.* **187,** 196 (1930)
51. R. Schoenheimer, S. Ratner, D. Rittenberg, *J. Biol. Chem.* **130,** 703 (1939)
52. R. F. Riley, *J. Am. Chem. Soc.* **66,** 512 (1944)
53. E. Chargaff, A. S. Keston, *J. Biol. Chem.* **134,** 515 (1940)
54. E. Chargaff, *J. Am. Chem. Soc.* **60,** 1700 (1938)
55. J. K. Parnas, *Enzymologia* **5,** 166 (1938)
56. E. L. Outhouse, *Biochem. J.* **31,** 1459 (1937)
57. G. Hevesy, *Advances in Enzymol.* **7,** 111 (1947)
58. C. K. Ingold, C. G. Raisin, C. L. Wilson, *J. Chem. Soc.* 915 (1936)

Scope of Health Physics

There has been no absolute definition of the scope of the proposed new subject of Health Physics, but in general it is concerned with the physics and biophysics involved in the interaction of radiation with the human body, with special emphasis on the protection of radiation workers against the potential hazards of their occupation. Health Physics is, thus, a restricted aspect of Medical Physics in which the basic intention can be seen from a study of proposed alternative titles. . . . Perhaps the only justification for regarding Health Physics as a new subject is that the recent large-scale expansion in the use of radiation and radioactive materials has apparently established the need for a group of men engaged full time in the considerations of radiation protection. In previous experience, medical physicists were required to give only part-time attention to the problems involved in their occupation. . . . Despite the fact that utilization of nuclear machines and other uses involving radiation and radioactive materials will undoubtedly expand during the next few decades, it is to be expected that the methods of hazard control will become sufficiently stabilized that specialists may no longer be required to regulate safety aspects, with the exception of the consultant services of a small group of qualified experts. This would restore the relative importance of the subject to its pre-war status.

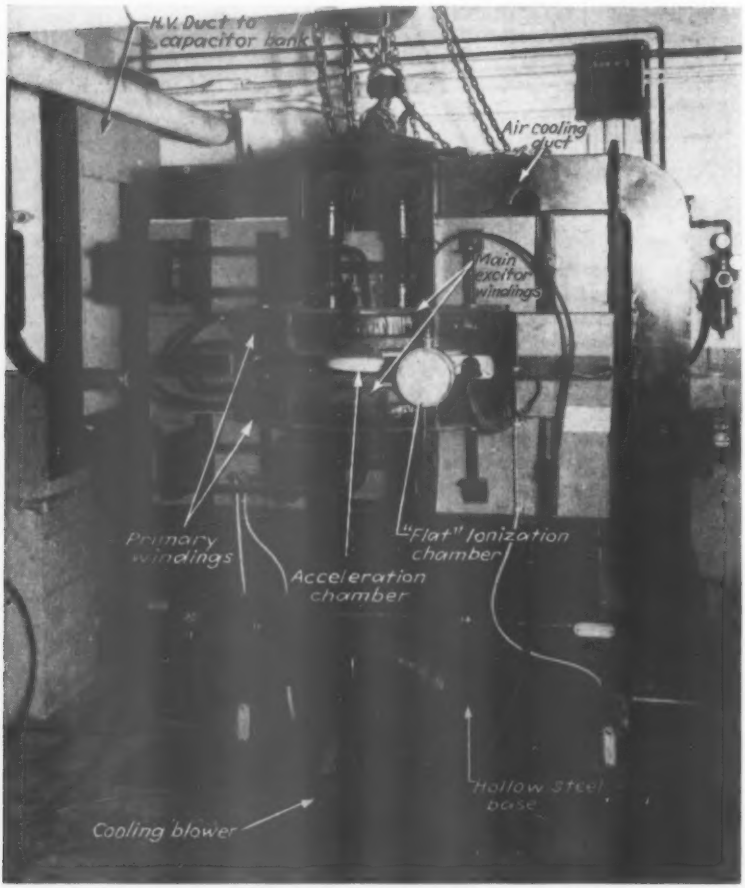
—MDDC 783, *H. M. Parker, Hanford Engineer Works*

THE NAVAL RESEARCH LABORATORY BETATRON*

BY

H. F. Kaiser, D. L. Mock, O. E. Berg, D. K. Stevens, and W. E. Harris

Naval Research Laboratory, Washington, D. C.



Complete assembly of betatron

Description of equipment and laboratory, featuring improvements introduced, such as the use of motor-generator drive for increased operational stability of the 20-Mev betatron. Basic betatron theory and use of the betatron in radiography and nuclear research are reviewed

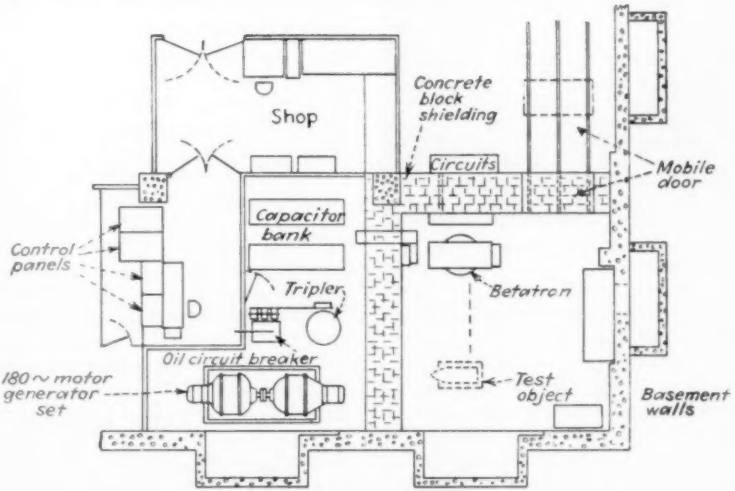


FIG. 1. Layout of betatron laboratory

DURING WORLD WAR II, the betatron became an interesting and promising device for both military inspection purposes and nuclear physics research. The National Defense Research Council promoted an intensive research and development program at the University of Illinois which had as its aim the utilization of the induction electron accelerator for research in government laboratories. The Naval Research Laboratory availed itself of these activities and in cooperation with the above-mentioned group built, during 1943 and 1944, a 20 Mev betatron and associated facilities incorporating features arising from the NDRC work and also some originated at the NRL. This unit was put into operation early in 1945 and has given satisfactory service. The details of this betatron laboratory are such that it may serve as an example for planning a 20-Mev betatron laboratory.

The general arrangement of the laboratory is shown in Fig. 1 which gives the floor plan of the research rooms. A number of locations were considered in choosing a site. Whereas a location in which the X-ray beam from the betatron could be directed above ground level would have been desirable, radiation protection requirements along with

* Condensed from NRL Report M-3014. Published by permission of Navy Dept.

FIG. 2. Electrically driven door for closing betatron room or for use as a baffle

other practical considerations indicated that a basement corner location would be more economical in providing radiation protection and in locating the betatron within reasonable distance of other research facilities.

In this particular building, advantage was taken of a light shaft outside the basement windows to provide a catching trap for the betatron beam. Steam, water and gas pipes present in the basement gave some problems but it was found possible to mount an I-beam with a 3½-ton chain hoist on the ceiling of the room with sufficient vertical clearance to perform all the necessary operations of assembling or disassembling the betatron yoke.

Inside radiation protection from the betatron is obtained from a 32-inch concrete wall, constructed (with staggered joints) from special solid concrete blocks made from 4:1 cement mixture. An opening of about 65 in. by 70 in. was left to accommodate an electrically driven mobile door, as shown in Figs. 1 and 2, rolling on a set of three flat rails embedded in the concrete floor. This door may be used either for completely closing the betatron room or as a baffle in case frequent access is wanted. The door consists of a hollow steel carriage frame within which concrete blocks are stacked, as in the barrier wall.

Radiation protection was found to be adequate outside of this room. Concern was had over the ceiling of the betatron room which was formed partly by large concrete-encased I-beams and partly by a four-inch concrete floor. The radiation measured in the room above indicated no serious intensities. This showed that the major portion of the betatron radiation is well directed.

A further consideration in the construction of the betatron laboratory was the sound produced by the various items of equipment. After the betatron had been put into operation, the Sound Division of the laboratory was asked to survey the acoustic conditions. In review at this point, it may be said that the noise level in the betatron room with the betatron operating at full capacity is about 97 decibels. Since personnel ordinarily cannot remain in the betatron room, the intense sound is not of serious consequence except for transmission effect on the rooms above or to the sides. Preliminary surveys also indicated a high and uncomfortable sound level (80 db overall) in the control room due to the motor-generator unit. The frequency spectrum of this sound pattern indicated sharp peaks at 30 and at 360 cycles at about 60–65 db sound intensities. Steps were taken to combat these conditions in the control room and motor-generator room, with the result that the sound level was reduced to 72 decibels overall in the control room with doors closed, a level that can be tolerated by the operators.

These acoustic findings are mentioned here briefly because they are an important consideration in any betatron

installation and especially so when the unit is to be used in hospitals or medical research laboratories. For these, it would be desirable to go a step further and completely enclose the betatron in an anti-resonant, sound-proof box specially designed for the type of sound generated by the unit. Betatrons operating at higher frequencies have an advantage that their sound is less unpleasant.

Fig. 1 shows, in addition to the features mentioned above, the disposition of the main components. A capacitor bank for resonant operation of the betatron is located near the betatron on the opposite side of the barrier wall. The position of the control room was chosen to be at the side of the betatron and at a reasonable distance away. The power sources and the oil circuit-breaker are located together with the capacitor bank, with convenient access from the control room.

Betatron Assembly

The basic theory and principles of operation for the betatron are discussed on p. 58. It is sufficient to say at present that the betatron is an electromagnetic device in which electrons are both accelerated and guided in an alternating magnetic field in such a way that they travel a circular path of substantially constant radius until their energy is converted in part to radiation by striking a target. This magnetic field is produced in a magnetic yoke similar to a transformer in construction in that large coils cause a magnetic flux to pass within them through the pole pieces and to circulate in a return path through the yoke. The complete betatron with the energizing coils and magnetic yoke apparent is shown on p. 46.

The yoke consists of bolted stacks of laminated iron spaced into parallel packages to give narrow slots for cooling. The yoke sits upon a hollow steel shell base from which air is drawn by the suction blower shown at the base of the unit. Air is thus drawn directly through the lower part of the yoke and likewise is drawn through the upper portion from a hood communicating with the base by a connecting duct at the side of the betatron. The base and hood have minor communicating ducts for cooling of the main coils.

Also shown is the power connection to the six-turn main energizing coils through four bakelite tubes. The top and bottom leads shown are connected to the portions of the top and bottom coils nearest the iron yoke and are connected together to ground at the side of the unit. The remaining two leads are connected to the leads communicating to the capacitor bank through the duct seen on the wall to the left. Here is also to be seen a pair of leads emerging from a bent conduit. These leads connect to the six-turn primary coil wound over the main energizing coils which are connected only to the capacitor bank. This primary coil is connected through the control stand to the power source (frequency tripler or motor-generator).

At the center of the yoke between the coils may be seen in part the evacuated porcelain acceleration chamber in which the electrons are accelerated. Radiation proceeds from a point target toward the right side and is directed out of the betatron in a conical bundle of radiation which passes through the circular shallow ionization chamber seen mounted directly on the front of the unit. The distribution and intensity of the radiation will be discussed further. See Fig. 3.

Fig. 4 shows a view of the porcelain accelerator chamber itself. This has two glass lead-in stems, the larger one serving as a glass base for the electron injector gun; the other for giving electrode connection to the internal metallized coating and vacuum "get-

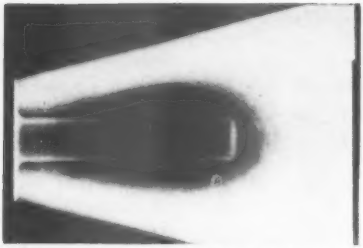


FIG. 3. Radiation from betatron

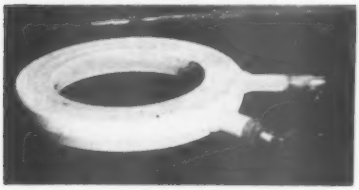


FIG. 4. Porcelain acceleration chamber

ters" and for connection to pumps in manufacture. This acceleration chamber is mounted in the betatron between specially shaped pole pieces which give the proper magnetic field to guide the electron in a circular orbit. The open space at the center of the acceleration chamber is filled, when mounted in the betatron, by a laminated iron centerpiece separated from the main pole pieces by short air gaps. This centerpiece serves to concentrate and carry the magnetic flux through the space encircled by the acceleration chamber. The change of this flux strength with time supplies the circular electric field (Faraday effect) that accelerates the electrons.

The electrons are supplied by a filament in an electron gun or injector but are introduced only for a brief instant of time as the magnetic field of the betatron is passing through its zero point. Then, electrons are injected into the acceleration chamber in a 2–8 microsecond pulse of 50–85 kv potential supplied by an r-f pulse transformer which forms part of the electron injector circuit. After injection and acceleration of the electrons during the quarter of a cycle increase in magnetic field, they are caused to strike a target on the injector gun by expansion of the orbit. This is accomplished in the 30 Mev unit by passing a heavy current through a separate single-turn coil cemented to each pole face immediately above and below the equilibrium orbit. This coil serves to expand the electron orbit by a sudden boost of the field. The current pulse is obtained by discharging a suitable capacitor through an ignitron into this coil at a desired instant in the quarter period. The pulse timing is varied by a phase-shifting device.

The timing of the injector pulse is obtained from a voltage pulse induced in a coil wound around a "peaker" strip of high permeability alloy. The timing of the injection pulse is also variable but only over a small time interval near the magnetic zero point. This is accomplished by purposely biasing the magnetization of the "peaker" strip with a direct-current winding also wound on this strip. The basic electron injection and orbit expansion circuits are shown in Fig. 5.

In addition to these controls on the electron beam, two additional circuits for improvement of X-ray output are used. One of these is merely a small, flat, trapezoidal coil operated at the same frequency as the main magnetic field and usually placed on the acceleration tube somewhere ahead of the electron gun to exert a focusing or steering action on the electrons. The other is a simple tube circuit which by discharge of a thyratron connects a short-circuiting coil, also cemented on the pole faces, which serves the purpose of purposely delaying the rise of magnetic field at the time of injection in the orbital region.

Control System

The above electronic circuits are controlled at the control stand on a panel

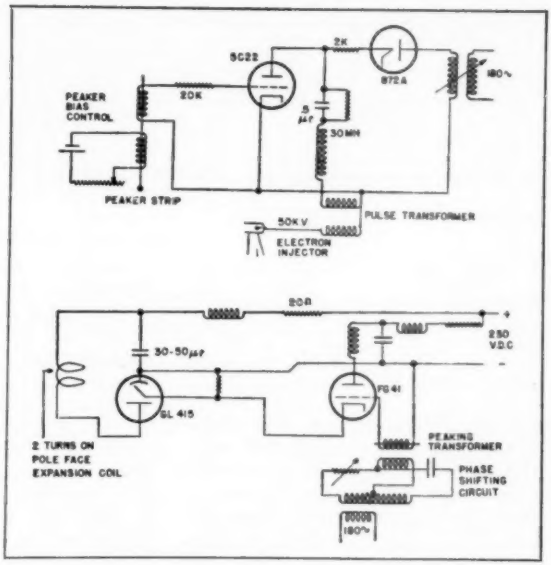


FIG. 5. Basic electron injection circuit (upper), basic electron orbit expansion circuit (lower)

immediately before the operator. A portion of these circuits are by necessity in the immediate vicinity of the betatron. Provision for occasional adjustment of these panels is made by mounting some of these circuits in cabinets on the wall of the betatron room on the side opposite the back of the unit. This facilitates the adjustment of these circuits while the unit is operating.

The control room and control panels are shown in Fig. 6. A diagram illustrating the relation of the control system to the betatron equipment is shown in Fig. 7. In Fig. 6, the two control boards on the right are the controls for the 75 kw, 180 cycle, Westinghouse motor-generator unit, while the two boards on the left are, respectively, the functional controls and the power metering and regulating board. The one at the extreme left has the necessary interlocked switches for controlling the separate operating circuits for the betatron such as vacuum

FIG. 6. Control panels

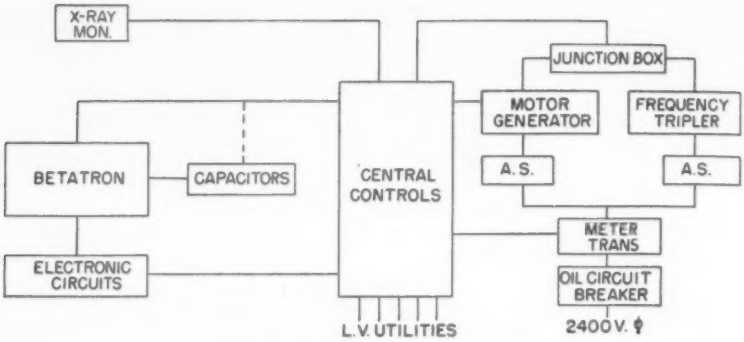


FIG. 7. Betatron electrical system

pumping, water cooling, air cooling, X-ray output metering, electron injection, orbit expansion, and the personnel warning system. The lower portion of the board contains also the indicating meters, including a cathode-ray oscillograph for indicating and checking the operation of these circuits. The second board has the meters for measuring the power input from the 2400 volt, three-phase power supply, the line frequency, voltage, and power factor; also meters for measuring the power generated by the 180 cycle power source as well as its voltage, current, and power factor. Direct indication of the last-mentioned quantity facilitates the tuning of the resonant driving circuit by giving a direct indication of the condition of the capacitor tuning.

The board also includes, in convenient position, a pair of rheostat controls for regulating the current in the 180 cycle power circuit and thus the amplitude of the current in the main driving circuit. A precision voltmeter on this panel is connected to two turns around the legs of the magnet yoke to indicate the amplitude of the magnetic flux change in the betatron. Since the electrons are accelerated in an orbit of substantially constant radius, this is also a measure of the energy of the electrons or X-rays produced, if the electrons are caused to expand into the target at maximum field amplitude. For expansion at other times, the energy is determined by that point in the quarter cycle at which the electron orbit expansion occurs. More recently, the upper left hand panel of the control stand has been fitted with small circuit-breakers and supply voltage indicating meters to eliminate fuse blocks in the low voltage utility lines.

As shown in Fig. 7, power is taken from a 2400-volt line cut into parallel connection with a line feeding the building. This is controlled by an oil circuit-breaker in the control room within easy reach of the operator in case a rapid cut-off of the power is required. This circuit-breaker also functions automatically to clear the line in case of capacitor cooling-water failure or in case of 110-volt board control voltage failure.

The motor-generator unit is a Westinghouse 93.8 kva 4 unit, 4-bearing, frequency charger unit consisting of a 125 hp, 2400 v, 3ϕ, 60 cycle, type 4-28-6G synchronous motor and 75 kw (80% P.F.) No. 12-28-10G single phase, 180 cycle, 1800 rpm, 12 pole, 240–480 v generator. The motor is started as an induction motor with

reduced voltage starting equipment.

The motor-generator unit or frequency tripler power supply is connected into the supply line as required by two sets of hook switches. To change from one type of power source to another requires, in addition, only a change in connection of the betatron primary coil circuit from one source to another within a junction box, an operation requiring only a few minutes.

Fig. 8 shows a group of driving circuits which were tried during the course of some experiments to determine the relative merits of tripler and motor-generator forms of drive for the betatron. The test results are shown in Fig. 9. These curves describe the change in magnetic amplitude, plotted as abscissa, observed when the line frequency changes by the amount indicated on the scale. The curves are labeled in groups A, B, C, indicating whether the betatron was excited (A) through its primary coupling winding, (B) through only its capacitor bank circuit on which a portion of the capacity is coupled to a high-voltage transformer inserted between the capacitor bank and power source, and (C) through the capacitor bank circuit again in such a way that the generator winding is in series with the main driving coils. These three systems of connection are shown in Fig. 8 and lettered correspondingly.

All curves with the exception of curve (1) were obtained using the motor-generator as the power source. These, with the exception of curve (2), show a smaller change of magnetic amplitude with a given change in line frequency. It was found that circuit of type A gave the best operating stability when driven by the motor-generator. The exceptional curve (2) was obtained by insertion of series capacitance in the primary circuit. The transformer coupling also appeared to be good as seen in curve (5). The results obtained with

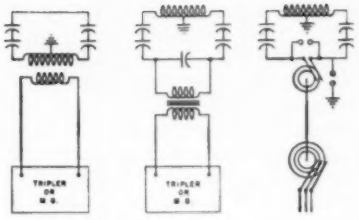


FIG. 8. Betatron drive circuits

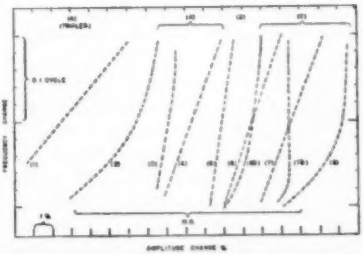


FIG. 9. Results of tests of motor-generator versus tripler drive

circuit (C) were not in general quite as good, but this particular system is well adapted to the heavy-current generator used and will be considered for further development and use.

In general, the results obtained in these tests indicated that the stability of operation using a motor-generator set was several times greater than that obtained with a frequency tripler of the type used. The relative cost of motor-generators and frequency triplers are not far enough apart to justify the preference of the tripler for economy. Although it is possible to stabilize by special electronic circuits or special circuit adjustment, the greater flexibility and control inherent in the m-g system will alone justify the choice.

Radiation Measurement

The radiation yield of the betatron is measured by ionization chambers which are connected to chassis and

output meters mounted on the control stand. Two "monitors" are used in this installation. One type is the circular "flat" monitor, which is seen on the front of the betatron on p. 46. This monitor is sensitive only to large radiation outputs such as obtained when the unit is functioning. Another monitor, also an ionization chamber of more conventional type but much more sensitive is used in conjunction with the shallow monitor. The more sensitive monitor chamber is located at a distance from the betatron and out of the direct beam, usually in the far corner of the betatron room near the ceiling. In the preliminary adjustment of the betatron, this monitor is brought near the unit and gradually removed as adjustment increases the yield of the unit. The two monitors and their circuits are shown in Figs. 10 and 11. Provision is made in the control stand to connect a remote Geiger counter to a vacuum tube voltmeter for cases in which very weak radiation is to be measured. This is desirable when the electron injection alone is being studied, the injected electrons being sufficiently energetic to produce weak X-rays.

To standardize the monitors, direct measurement of the radiation output is made with a 25 r thimble ionization chamber which is inserted in a bakelite well (shield) having a wall thickness of about 5 cm. The latter serves to shield the ionization chamber from strong electrons and to set up an equilibrium between the energetic X-rays and the electrons they are constantly generating in passage through the air. This procedure is necessary to obtain a reliable measure of the radiation output. The output monitors are calibrated in terms of these chamber measurements and in general it will be found that a separate calibration is required for each Mev maximum electron energy. Near maximum amplitude, the roentgen-sensitivity of the monitor to maximum electron energy varies a few percent so the panel meter readings serve to give a fairly close indication of the "r" output per minute and serve to determine the time of exposure to be used in radiography.

Operation Procedure

The radiation monitors determine the adjustments made by the operator in operation of the betatron. In brief, the operation of the betatron is not as formidable as it might appear and consists in the following series of operations:

a. Energizing the control stand which can be so set that a single switch will energize all function units providing each of these has been switched on with its individual unit switch (to facilitate "trouble shooting," each function unit may be separately energized).

b. Switching on of the 2400-volt power mains through the main circuit breaker.

c. Starting of motor-generator unit through its reduced voltage starting gear and adjustment of generator voltage and power-factor of synchronous motor operation.

d. After time delay relays have closed (permitting vacuum tube pre-warming), attention is then directed to the electron injection and orbit central function units. After the orbit expansion unit has been checked for operation, the injector filament and plate voltages are set to required values.

e. The magnet is energized by the motor-generator load switch, or by the control stand starting switch, and the magnetic amplitude set with rheostat controls to a desired value as given on the precision voltmeter.

f. The orbit expander circuit is switched on. The injector bias current is then adjusted and if all else is in order, an X-ray output is observed on the monitor meters when the bias current reaches a correct value.

g. Final adjustments are made on

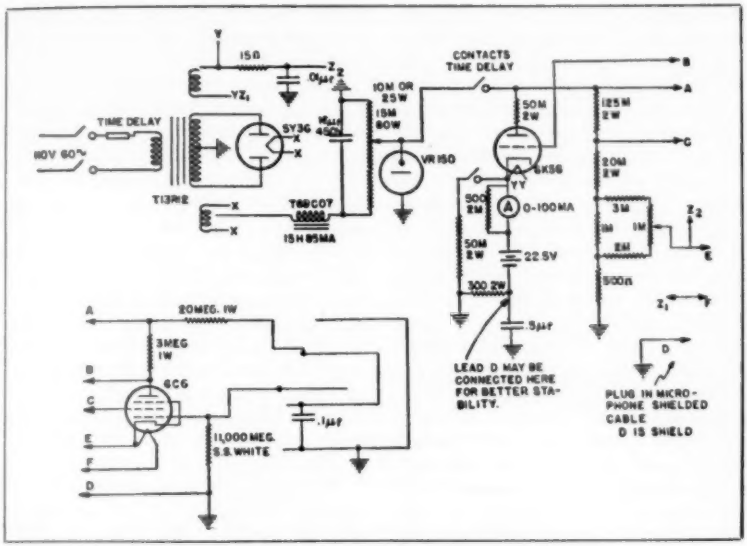


FIG. 10. Sensitive monitor and ionization chamber

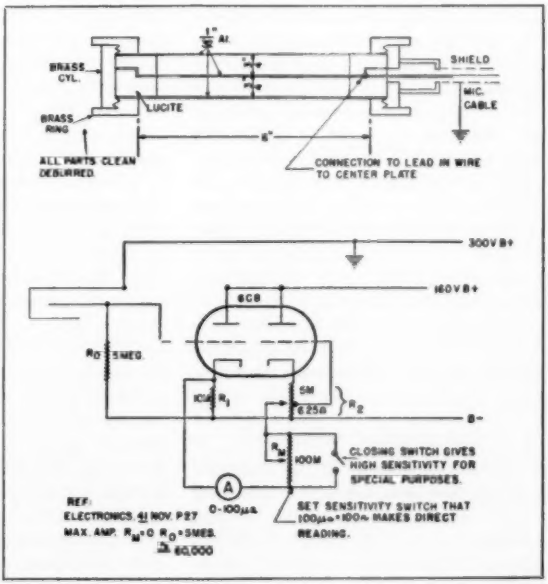


FIG. 11. "Flat" ionization chamber

the injection and expansion controls including such controls as the booster circuits to obtain best yields possible for the given magnetic amplitude.

After these operations are completed, the unit may be allowed to run continuously and X-rays turned on or off by switching the expander in and out of action. Continual adjustment of the controls is not required when using the motor-generator unit as power source. The equipment will operate "hands-off" for considerable periods before correction adjustments are required. When the electron injection and orbit expansion circuits have been developed to the point of freedom from occasional breakdown and erratic operation, the operation of large betatrons will be almost as simple and convenient as the operation of any industrial X-ray equipment.

The cost of operation of a betatron varies of course with size. Power consumption is generally higher than other forms of X-ray generators and is particularly higher in betatron operation at high driving frequency. The above-described 20 Mev unit usually operates at less than 35 kw power of which 15–20 kw represents the losses in the betatron itself. The remainder represents the power consumption and losses of auxiliary equipment. At power supply rate, this item of expense is not excessive in view of work capability and relative cost of photographic materials used. A larger expense item is maintenance. This will be equivalent to one man's full time activity for a 20 Mev installation. In an industrial installation, the maintenance man may also be the operator.

Radiographic Applications

The Naval Research Laboratory's betatron has been used for the radiography of various foundry castings (ranging from 4 to 8 inches in thickness), various electrical and optical equipment, and explosives. The largest item radiographed has been a 15-inch, high-explosive shell.

The betatron radiation has a distinct advantage in the fineness of its source. The natural width of the betatron beam in the vertical direction is a small fraction of a millimeter—0.05 inches. This beam, in expanding into a platinum target on the injector, causes a focal spot of the above height and a width that is about negligible due to the small pitch of the spiral in which the electrons approach the target and is about 0.005 inches. To improve the fineness, the target is made of platinum in the shape of a wedge so that it will plow into the beam as the latter expands. The vertical dimension of the focal spot is thereby reduced.

The fine focus allows certain radiographic techniques to be employed. One is the magnifying technique in which the test object is placed close to the betatron and the film cassette at a greater distance. Considerable magnification without loss of detail is possible. Another technique is the close duplication of dimensions at longer film-focal distances with allowances made for known film shrinkage to allow working drawings or measurements to be made directly.

The distribution of the intensity of the radiation from the betatron is not uniform but is distributed across the diameter of a narrow cone as shown in Fig. 12. This distribution has to be taken into account in radiography and, while a heavily exposed central spot is often objectionable on a radiograph, advantage can often be taken of the intensity distribution to radiograph a considerable range of thicknesses in a simple exposure. The central spot effect may be minimized either by using a large film focal distance or by setting the test object so that the intense central spot does not fall on the radiograph. The disadvantage may also be removed

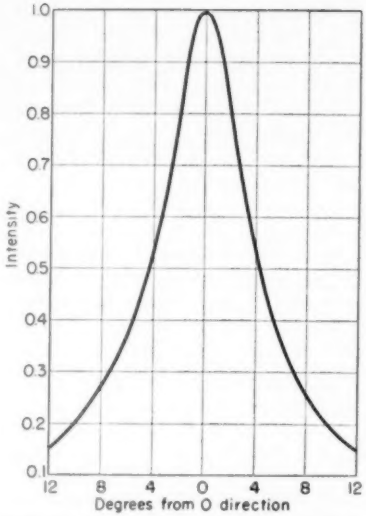


FIG. 12. Distribution of X-ray intensity in beam at 19 Mev

by use of an artificial diaphragm plate of proper shape and range of thicknesses to compensate the intensity distribution.

It should also be mentioned at this point that due to the intense forward scattering characteristics, little provision is required for "blocking" the test object as in low-voltage radiography. A figure of merit which gives the ratio of the film blackening due to scattering to that due to the direct primary radiation is usually 6–8 for 1 Mev radiation but becomes quite small (less than 1) for betatron radiation.

Application to Nuclear Technique

The betatron has potentialities in several directions that have only been explored in part. Radiation of high energy has interesting properties in that it is able to bring about such nuclear effects as electron pair production and photo-nuclear reactions in which a quantum of gamma radiation is absorbed in the nucleus and a reaction results in which electrons, protons or neutrons are ejected. A considerable number of such reactions have been studied but these represent only a fraction of those possible. As the betatron energy is increased, these processes become more complicated and even photo-fission of the nucleus may be observed. It is believed that mesotrons may also be produced although such effects are to be expected mainly at energies somewhat above 100 Mev.

Artificial radioactivity may be induced in a number of substances by irradiation with 20 Mev radiation, copper being an example. These activities are usually of positron emitting types and are not very intense. The betatron does not compare or compete with the cyclotron in this respect. These induced radioactivities however may be used to advantage in various forms of experimental research. Since each substance has a threshold Mev it is possible to use this fact for determination of absolute absorption coefficients or for identification in some cases.

With a special acceleration chamber, the betatron may induce much higher artificial radioactivity by introducing the test material as a target into the circulating electron beam. Neutrons may also be produced directly by using a beryllium target. It has recently become possible, by means of a special magnetic shunt, to extract the electrons from the betatron upon expansion of the electron orbit. Many interesting studies can be made using these high energy cathode rays, which are able to penetrate considerable thickness of material (several cm depending on energy and absorber). The use of these is expected to be particularly valuable in the field of medicine where the therapeutic depth dose will then be localized near the end of the range of electrons in flesh.

The use of the betatron as a neutron source offers another line of experi-

mentation which should be kept in mind. Since the radiation output of the betatron at 50 r per min at one meter is 3600 gm-radium equivalent, much stronger neutron sources than obtainable by radium sources are now available. This gives a neutron flux of at least 3.6×10^6 neutrons per second. This is small compared to the neutron flux obtained in uranium piles where a flux of about 10^8 neutrons per sq. cm per kw may be produced. Radiography with neutrons has been contemplated for some years but no particular success has yet been published. The interesting properties of neutrons, however, make further study attractive, and, in the field of metallurgical research, the effects of neutrons in promoting "dislocations" in metals is an interesting topic.

APPENDIX

Basic Theory of Betatron

The operation of the betatron depends primarily on the presence of a circular electric field which links a magnetic flux changing with time. With suitable magnetic conditions, the particle will be guided in a stable path of substantially constant radius while under the influence of this electric field. These are the basic factors involved.

The electric field in this circular path is easily obtained since we know the expression for the emf (or total work) induced in a loop enclosing the changing flux. If this is V, the field

$$E = \frac{V}{2\pi r} = \frac{-1}{2\pi r}\frac{d\phi}{dt} \qquad (1)$$

The sign of the induced voltage is important when the nature of the charge is to be considered because it determines the direction, with respect to the field, in which it will be possible to drive the particle. Noting again that $V = -d\phi/dt$, the convention expressed is that with a *decreasing* field the electric field will be such that a *positive* particle will be driven in a *clockwise* direction when *looking in the direction* of the magnetic field. For calculation of relations between forces, the sign need not be considered.

The electric field E exerts on a charge e a force

$$F = \frac{Ee}{c} \qquad (2)$$

The quantity c represents the ratio between the numerical value of charge expressed in electrostatic and electromagnetic units, respectively ($c = 3 \times 10^{10}$ cm per sec). From mechanics (Newton's law), this force must also be given by $F = \frac{d}{dt}(mv)$ or simply p';

$$\text{then} \quad F = \frac{Ee}{c} = p' = \frac{e}{2\pi rc}\phi' \qquad (3)$$

Next it is necessary to examine the relation between the centrifugal force which the particle of mass m will exert when it is constrained to move in a circular path of radius r to the necessary magnetic force which is required to balance this force. The mechanical force is mv^2/r. It is known that a charged particle moving in a direction at right angles to a field of constant intensity will have a magnetic force upon it that is at right angles to both the field and the direction of motion, with the result that it travels in a circle.

The magnetic force follows a vector law $\mathbf{F}_m = \mathbf{I} \times \mathbf{H}$ and for the present purpose we may convert this to a scalar equation $F = Hev$ where the current I is now represented by a charge e times its velocity. This force must be exactly equal to the mechanical centrifugal force to cause motion in a circle or

$$\frac{mv^2}{r} = \frac{Hev}{c} \text{ (esu)} \qquad (4)$$

$$\text{or } mv = \frac{Her}{c} = p, \text{ the momentum} \qquad (5)$$

The time derivative of this quantity has already been determined above, so by differentiation of (5),

$$p' = \frac{d}{dt}\left(\frac{Her}{c}\right) = \frac{e}{2\pi rc}\frac{d\phi}{dt} \qquad (6)$$

and integrating $\frac{Hr}{c} = \frac{1}{2\pi rc}\phi + \text{const.}$

$$2\pi r^2 H = \phi - \phi_0 \qquad (7)$$

where ϕ_0 represents the constant of integration and is easily seen to be any constant magnetic flux superposed on the given flux. This equation, known as the 1:2 rule, states that the magnetic field existing in the region where the electron is being guided in a circular path

$$H = \frac{1}{2}\left(\frac{\phi - \phi_0}{\pi r^2}\right) = \frac{1}{2}\overline{H} \qquad (8)$$

is thus such that it is one half the average field strength computed from the total flux linked by the electron orbit.

Another more advanced way of looking at this is to let the current (electron beam) have an associated vector potential $A\phi$. Now the flux through the circular orbit is very generally $\phi = \int Hds = \int \text{curl}\, A_\phi ds$ which by Stokes law (which says that the surface integral of the curl of a vector may be replaced by the line integral of that vector around any closed path representing that surface) is now

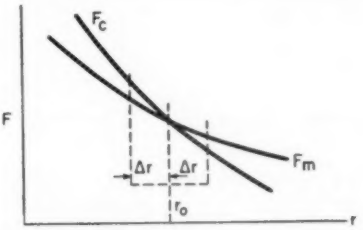
$$\phi = \int A_\phi dl = 2\pi r A_\phi \qquad (9)$$

since the path of integration is the circular path. The field is given by $H_z = \text{curl}_z A_\phi$ or

$$H_z = \frac{\partial A_\phi}{\partial r} + \frac{1}{r}A_\phi + r\frac{\partial A}{\partial \phi} \qquad (10)$$

Since the field is circularly symmetric $\frac{\partial A_\phi}{\partial \phi} = 0$ and since the vector potential has a minimum at the equilibrium position $\frac{\partial A_\phi}{\partial r} = 0$ leaving $A_\phi = rH_z$ and then $\phi = 2\pi r A_\phi = 2\pi r^2 H_z$ which again is the 1:2 rule. An added constant ϕ_0 can be introduced into this expression also.

The conditions for a radially stable orbit may now be examined again with equation (4) and the two parts of this important equation are plotted in curve form in the figure below where the curve shows the centrifugal force decreasing as $1/r$ and the other, the magnetic force Hev, decreasing in a manner determined by the rate of decrease of H. At some



radius r_0, these are equal and it is seen that on either side of this point Δr, the magnetic force, is greater or less than the centrifugal force. In such cases

$$\frac{mv^2}{r_0 + \Delta r} = \frac{mv^2}{r_0\left(1 + \frac{\Delta r}{r}\right)}$$

$$= \frac{mv^2}{r_0}\left(1 - \frac{\Delta r}{r}\right) \text{ approximately.} \qquad (11)$$

The magnetic field may be assumed to vary according to some power law

$$H = H_0\left(\frac{r_0}{r}\right)^n = H_0\left(\frac{r - \Delta r}{r}\right)^n \qquad (12)$$

$$H = H_0\left(1 - \frac{\Delta r}{r}\right)^n$$

$$= H_0\left(1 - n\frac{\Delta r}{r}\right) \text{ approximately.}$$

Let F_0 represent the existing centrifugal (or centripetal) force at the balance point. The modified forces then may be written in the forms

$$F_0 = \left(1 - \frac{\Delta r}{r}\right) = F_c \qquad (13)$$

$$F_0 = \left(1 - n\frac{\Delta r}{r}\right) = F_m \qquad (14)$$

The radial force restoring the electron to its equilibrium point is the difference $F_m - F_c = -F_0(n-1)\frac{\Delta r}{r}$ (ap-

proximately). This force resembles the force of an elastic spring and causes a vibration of the electron about its equilibrium radius. From Newton's law this is equal to the radial time rate of change of momentum and gives a differential equation of the form,

$$m\frac{d^2\Delta r}{dt^2} + F_0(n-1)\frac{\Delta r}{r_0} = 0$$

$$\frac{d^2\Delta r}{dt^2} + \frac{v^2}{r_0^2}(n-1)\Delta r = 0 \quad (15)$$

of which the solution

$$\Delta r = \Delta r \cos(\omega_0\sqrt{1-n})t$$

is a simple harmonic motion. Actually the situation is more complicated and it is necessary to consider the relativistic change in mass of the accelerated particle as it is being accelerated.

From equation (15) the restoring force constant is seen to be $\frac{v^2}{r_0^2}(n-1)$. The potential energy of a displacement Δr is $\frac{1}{2}K(\Delta r)^2$ and this will represent to good approximation the kinetic energy of transverse vibration of the electron which moreover may be assumed to remain constant as the electron is accelerated in its orbit. Then

$$W = \frac{1}{2}\frac{v^2}{r_0^2}(n-1)(\Delta r)^2 = E \quad (16a)$$

or $$(\Delta r)^2 = \left[\frac{2Wr_0^2}{(n-1)}\right]\frac{1}{v^2} = c\frac{1}{v^2} \quad (16)$$

Differentiating in this form,

$$2(\Delta r)d(\Delta r) = -2\frac{c}{v_3}dv \quad (17)$$

which, on dividing by (16), gives

$$\frac{d(\Delta r)}{\Delta r} = -\frac{dv}{v} = -\frac{1}{2}\frac{dE}{E} \quad (18)$$

Since holding Δr as fixed for a moment, equation (16a) may be written as $W = E = \text{const.} \times v^2$ from which

$$\frac{dW}{W} = \frac{dE}{E} = \frac{2dv}{v}.$$

The vibration in the vertical or z direction also obeys an equation similar to (15). The magnetic restoring force is $H_r ev$. The curl of the magnetic field is zero and gives $H_r = -nH_z\frac{z}{r}$

so $$H_r ev = -\frac{nz}{r}$$

and $$(H_z ev) = -\frac{nz}{r}F_0 \quad (19)$$

Then
$$\frac{d^2z}{dt^2} - F_0\frac{nz}{r} = 0$$

$$\frac{d^2z}{dt^2} - \frac{v^2}{r_0^2}\frac{nz}{r} = 0 \quad (20)$$

which by analogy with equation (15) has a solution $z = z_n \cos(\omega_0\sqrt{n})t$.

Recapitulating, the electron has a radial restoring force which varies as $(1-n)$ and a radial vibration frequency varying as $\sqrt{1-n}$; also vertical restoring forces and vibration frequency varying as n and $\sqrt{n}$, respectively. The cross section of the electron beam is thus elliptical with axial ratio given by $\sqrt{\frac{1-n}{n}}$.

The above has covered the magnetic flux conditions and the stability of the electron orbit. It is necessary yet to state the energy of the electron. The relativistic formula

$$E = m_0c^2\left[\frac{1}{\sqrt{1-B^2}} - 1\right]$$

together with equation (6) gives

$$E = 3\times10^{-4}\left(\frac{Hr}{B}\right) - 0.51 \text{ (Mev)} \quad (22)$$

when H is expressed in gauss and r in cm.

The main points of basic betatron theory have been given above in elementary form. There are other technical considerations which are beyond the scope of the appendix, but some of these may be mentioned. These include (*a*) the effect of space charge in spreading electrons in the orbit and its neutralization at relativistic speeds by the parallel current attraction forces, (*b*) the scattering of electrons by gas molecules, (*c*) the technique of injecting electrons and the oscillations produced in this process, and (*d*) positive ion space charge neutralization.

BIBLIOGRAPHY ON INDUCTION ELECTRON ACCELERATORS

1. J. Slepian. X-ray Tube. U. S. Patent 1,645,304 (1927)
2. G. Breit, M. A. Tuve, O. Dahl. *Carnegie Inst. Yearbook* 1927–1928, p. 209
3. R. Wideröe. On a New Principle for Generation of High Voltages. *Arch. Elektrotech.* **21,** 387 (1928)
4. E. T. S. Walton. The Production of High-speed Electrons by Indirect Means. *Proc. Cambridge Phil. Soc.* **25,** 469 (1929)
5. M. Steenbeck. Device for Producing Electron Rays of High Energy. U. S. Patent 2,103,303 (1937). DRP (656,378-698,867)
6. W. W. Jassinsky. Acceleration of Electrons in an Alternating Electromagnetic Field. *Arch. Elektrotech.* **30,** 590 (1936); *J. Exp. Theoret. Phys.* U.S.S.R. **5,** 983 (1935); *Electricity* U.S.S.R. **3,** 37 (1936)
7. G. W. Penney. U. S. Patent 2,193,602 (1940)
8. Ia. P. Terletsky. Relativistic Problem of Movement of an Electron in an Alternating Magnetic Field. *J. Exp. Theoret. Phys.* U.S.S.R. **11,** 96 (1941)
9. D. W. Kerst. Acceleration of Electrons by Magnetic Induction. *Phys. Rev.* **58,** 841 (1940); *Phys. Rev.* **59,** 110 (1941); *Phys. Rev.* **60,** 47 (1941)
10. D. W. Kerst, R. Serber. Electron Orbits in Induction Accelerator. *Phys. Rev.* **60,** 53 (1941)
11. D. W. Kerst. Twenty Million Electron Volt Betatron or Induction Accelerator. *Rev. Sci. Instr.* **13,** 387 (1942)
12. D. W. Kerst. Magnetic Induction Accelerator. U. S. Patent 2,297,305 (1942)
13. M. Steenbeck. Acceleration of Electrons by Alternating Fields. *Naturwissenschaften* **31,** 234 (1943)
14. G. C. Baldwin. Magnetic Induction Accelerator" U. S. Patent 2,331,788 (1943)
15. A. R. Wildhagen. The Betatron. *Scientific American,* May, 1943
16. Research Note on 100-Mev Betatron *Gen. Elec. Rev.* **46,** 58 (1943); *Science News Letter* **48,** 278 (1945)
17. D. W. Kerst. The Betatron. *Radiology* **40,** 115 (1943)
18. D. W. Kerst. Electron Induction Accelerator. *Ind. Radiography* **3,** 36 (1944)
19. D. W. Kerst. The Betatron. *Electronic Eng.* **16,** 493 (1944); *Ind. Radiography* **5,** 45 (1946)
20. T. J. Wang. The Betatron. *Electronics* **18,** June 1945
21. E. E. Charlton, W. F. Westendorf. 100 Million Volt Induction Electron Accelerator. *J. Applied Phys.* **16,** 657 (1945)

Flux Bias Betatrons

22. D. W. Kerst. Method of Increasing Betatron Energy. *Phys. Rev.* **68,** 233 (1945)
23. W. F. Westendorf. Use of Direct Current in Induction Electron Accelerators. *J. Applied Phys.,* **16,** 657 (1945)

See also References 34, 44 and 45.

Electron Radiation Losses

24. D. Iwanenko, A. Pomerantschuk. On the Maximal Energy Obtainable in a Betatron. *Compt. rend. acad. sci.* (U.S.S.R.) **44,** 315 (1944); *Phys. Rev.* **65,** 343 (1944)
25. J. P. Blewett. Radiation Losses in the Electron Accelerator. *Phys. Rev.* **69,** 87 (1946)
26. L. I. Schiff. Production of Particle Energies beyond 200 Mev. *Rev. Sci. Instr.* **17,** 6 (1946)
27. J. Terletsky. On the Stability of the Electron Motion in the Induction Accelerator. *J. Phys.* (U.S.S.R.) **9,** 159 (1945)

Radiography

28. G. D. Adams. Radiography with the Betatron. *Ind. Radiography* **4,** 23 (1945–46)
29. J. P. Girard, G. D. Adams. Applications of the Betatron to Practical Radiography. *Elec. Eng.* **65,** 241 (1946)
30. G. M. Almy, G. D. Adams. Three to Twenty Million Volt Radiography. OSRD Final Report No. 5067 (contains bibliography of 12 reports and inventions on NDRC Betatron Project)

General

31. D. W. Kerst. Historical Development of the Betatron. *Nature* **157,** 90 (1946)
32. H. C. Pollock. The Beta-synchrotron. *Phys. Rev.* **69,** 125 (1946)
33. H. F. Kaiser, E. C. Greanias. The Synchro-betatron, Electron Accelerator Guide Fields. *Phys. Rev.* **69,** 536 (1946)
34. E. Amaldi, E. Ferretti. On Two Possible Modifications of the Induction Accelerator. *Rev. Sci. Instr.* **23,** 277 (1946)
35. I. Paul, T. J. Wang. Betatron Pulsing System, *Electronics* **19,** 156 (1946)
36. W. Bosley. The Betatron. *J. Sci. Instr.* **23,** 277 (1946) (Metropolitan Vickers 20 Mev)
37. Anon. The Betatron. *Beama J.* **53,** 306 (1946) (English 150-ton betatron)
38. T. J. Wang. On Increasing the Effectiveness of a Betatron. *Phys. Rev.* **69,** 42 (1946)

Electron Beam Extraction

39. L. S. Skaggs, G. M. Almy, D. W. Kerst, L. H. Lanzl. Removal of the Electron Beam from the Betatron. *Phys. Rev.* **70,** 95 (1946)
40. E. D. Courant, H. Bethe. On Bringing Beam out of a Betatron. *Phys. Rev.* **70,** 798 (1946)
41. J. S. Clark, I. A. Getting, J. E. Thomas, Jr. A New Method for Displacing the Electron Beam in a Synchrotron. *Phys. Rev.* **70,** 562 (1946)
42. R. Wideröe. On Bringing Beam out of a Betatron. *Phys. Rev.* **71,** 376 (1947)
43. H. W. Koch, D. W. Kerst. Note on Methods of Displacing the High Energy Beam in a Betatron. *Rev. Sci. Instr.* **18,** 681 (1947)

General

44. H. F. Kaiser. European Electron Induction Accelerators. *J. Applied Phys.* **18,** 1 (1947)
45. Kollath, Widerõe, and Tounchek. Betatron Papers and General Information on Megavolt Forshungsanstalt (MVF). U. S. Dept. Commerce Pub. Bd. Items 2197, 2198, and 2199.
46. Siemens-Reiniger. 5–6 Mev Betatrons. U. S. Dept. Commerce Pub. Bd. Item 2201
47. R. Widerõe. Effect of Phase Displacement between the Inducing and Guiding Fields of a Betatron. *Rev. Sci. Instr.* **18,** 250 (1947)
48. J. Rachmann, E. Cherry. Electron Mechanics of Induction Acceleration (Parts I and II). *J. Franklin Inst.* **243,** 261, 345 (1947)
49. I. Kaestle. Power Supply for 100-Mev Betatron. *Elec. World* **128,** No. 9, 42 (1947)
50. E. C. Gregg Jr. A. C. Probe for Measurement of Magnetic Fields (Betatron). *Rev. Sci. Instr.* **18,** 77 (1947)
51. W. B. Lasich. Note on 2.8 Mev Betatron. *J. Sci. Instr.* **24,** 177 (1947)
52. J. P. Blewett. Magnetic Field Configurations Due to Air Core Coils. *J. Applied Phys.* **18,** 968 (1947)
53. J. P. Blewett. Design of an Air Core Synchrotron. *J. Applied Phys.* **18,** 976 (1947)
54. G. D. Adams, D. W. Kerst, D. T. Scag. Azimuthal Variations of Betatron Fields at Injection Time. *Rev. Sci. Instr.* **18,** 799 (1947)
55. E. C. Crittenden, W. E. Parkins. Methods for Betatron or Synchrotron Beam Removal. *J. Applied Phys.* **17,** 444 (1946)
56. W. E. Parkins, E. C. Crittenden. A Graphical Method of Determining Particle Trajectories. *J. Applied Phys.* **17,** 447 (1946)
57. D. DePackh, A. V. Haeff. An On-orbit Injector for Betatrons and Synchrotrons. *Phys. Rev.* **72,** 247 (1947)
58. G. C. Baldwin, G. S. Klaiber, A. J. Hartzler. Synchronization of Auxiliary Apparatus with a Betatron. *Rev. Sci. Instr.* **18,** 121 (1947)
59. H. F. Kaiser. Remote Control Betatron Target Positioner. *Rev. Sci. Instr.* **18,** 134 (1947)

Research with Betatron

60. W. H. Koch, D. W. Kerst, P. Morrison. Experimental Depth Dose for 5, 10, 15 and 20 Million Volt X-rays. *Radiology* **40,** 120 (1943)
61. J. D. Cockroft. The Cyclotron and Betatron. *J. Sci. Instr.* **21,** 189 (1944)
62. Anon. Betatron for Cancer Research. *Science News Letter* **48,** 373 (1945)
63. G. C. Baldwin, H. W. Koch. Measurements of the Threshold Photo-Nuclear Effect. *Phys. Rev.* **68,** 1 (1945)
64. E. E. Charlton, G. C. Baldwin. Nuclear Research with the 100-Mev Betatron. *Proc. Natl. Electronics Conf.* **2,** 650 (1946)
65. G. C. Baldwin, G. S. Klaiber. Multiple Nuclear Disintegrations Induced by 100 Mev X-rays. *Phys. Rev.* **70,** 259 (1946)
66. M. Schein, A. J. Hartzler, G. S. Klaiber. Production of Heavily Ionizing Particles by X-rays Generated by 100 Mev Betatron. *Phys. Rev.* **70,** 435 (1946)
67. G. S. Klaiber, A. E. Luebke, G. C. Baldwin. Range-Momentum Measurement of Particles Emitted in Nuclear Disintegrations Induced by 100-Mev X-rays. *Phys. Rev.* **70,** 789 (1946)
68. D. W. Kerst. Development of the Betatron and Application of High Energy Betatron Radiations. *Am. Scientist* **35,** 57 (1947)
69. J. A. Wheeler. Elementary Particle Physics. *Am. Scientist.* **35,** 177 (1947)

"Once the engineer was an artisan. He rose from the ranks; but there has been a change in the past 40 years. Germany was the first nation to recognize the necessity of a deeper scientific approach to engineering. She began to develop a group of engineers, professors and research men and to educate them in basic science as underlying the development of technology. For she recognized the importance and necessity of a basic and scientific foundation for technology and engineering development. This was largely responsible for Germany's strength in the last two wars."

—Boris A. Bakhmeteff, on being awarded the Norman Medal by the American Society of Civil Engineers

BIBLIOGRAPHY

RADIATION MEASUREMENT

By MONICA HEALEA

Department of Physics, Vassar College, Poughkeepsie, New York

THIS BIBLIOGRAPHY, the first part of which was published in the December NUCLEONICS, was prepared for the Electronics Division at Brookhaven National Laboratory. The portion appearing below covers the subjects of ionization chambers, electrometers, other counting methods, and statistics. References to other subjects relating to radiation measurement will be published next month.

IONIZATION CHAMBERS

A. H. COMPTON, E. O. WOLLAN, R. D. BENNETT. A Precision Recording Cosmic-Ray Meter. *Rev. Sci. Instr.* **5,** 415 (1934)

LEE A. DUBRIDGE, HART BROWN. An Improved D. C. Amplifying Circuit. *Rev. Sci. Instr.* **4,** 532 (1935)

GLADYS D. FINNEY, ROBLEY D. EVANS. The Radioactivity of Solids Determined by Alpha-Ray Counting. *Phys. Rev.* **48,** 503 (1935)

I. FRANK. Compensation Chamber for Analyzing the Composition of Radioactive Preparations by their Gamma-ray Activity. *J. Phys. U.S.S.R.* **9,** 123 (1945)

CARL T. HIPDON, M. L. POOL, J. D. KURBATOV. Radioactive Scandium I. *Phys. Rev.* **67,** 289 (1945)

LYNN G. HOWELL. Gamma-Ray Measurements in Oil Wells. *Electronics* **17.3,** 134 (1944)

G. C. LAURENCE. Measurement of Extra Hard X-Rays and Gamma-Rays in Roentgens. *Can. J. Research* **A15,** 67 (1937)

LLOYD G. LEWIS. Study of Cosmic Ray Air Showers with the Method of Coincident Bursts in Two Unshielded Ionization Chambers. *Phys. Rev.* **67,** 228 (1945)

D. MAEDER. Electron-Tube Spectrograph for the Measurement of Ionization Chamber Pulses. *Helv. Phys. Acta.* **20,** 2, 139 (1947)

ROBERT A. MILLIKAN, H. VICTOR NEBER. A Precision World Survey of Sea-Level Cosmic-Ray Intensities. *Phys. Rev.* **50,** 15 (1936)

C. G. MONTGOMERY, D. D. MONTGOMERY, W. E. RAMSEY, W. F. G. SWANN. A Search for Protons in the Primary Cosmic-Ray Beam. *Phys. Rev.* **50,** 403 (1936)

GUSTAV ORTNER, GEORG STETTER. A Method for the Measurement of Strong Polonium Preparations. *Physik Z.* **35,** 563 (1934)

E. RUTHERFORD, F. A. B. WARD, C. E. WYNN-WILLIAMS. A New Method of Analysis of Groups of Alpha-Rays-(1), The Alpha Rays from Radium C, Thorium C and Actinium C. *Proc. Roy. Soc. London* **A129,** 211 (1930)

RUBBY SHERR, ROLF PETERSON. Study of Pulse Shapes in a Parallel Plate Ionization Chamber. *Rev. Sci. Instr.* **18,** 567 (1947).

B. S. SMITH, H. CARMICHAEL, J. F. STELJES, C. O. PEABODY, D. S. CRAIG. Design and Performance of Two of the Chalk River Ion-Chambers. *Bull. Am. Phys. Soc.* **22,** No. 3, 8 (1947)

W. F. G. SWANN. Detection of the Ionization by Individual Cosmic Rays. *Phys. Rev.* **43,** 775A (1933)

W. F. G. SWANN, W. E. RAMSAY. Ionization Spurts Resulting from Cosmic Ray Entities. *Phys. Rev.* **45,** 758A (1934)

WILLIAM D. URRY. Ionization Chamber for Counting Alpha Particles. *Rev. Sci. Instr.* **12,** 289 (1941)

E. WILHELMY. Resonance Groups of Protons and Alpha-Particles Produced by Neutron Bombardment. *Z. Physik* **107,** 769 (1937)

CHIEN-SHUING WU, EMILIE SEGRE. Radioactive Xenons. *Phys. Rev.* **67,** 142 (1945)

C. E. WYNN-WILLIAMS, F. A. B. WARD. Valve Methods of Recording Single Alpha-Particles in the Presence of Powerful Ionizing Radiations. *Proc. Roy. Soc. London* **A131,** 391 (1931)

HERBERT IVAN ZAGOR, FRANK A. VALENTE. Heavy Particle Groups from the Neutron Disintegrations of Nitrogen and Neon. *Phys. Rev.* **67,** 133 (1945)

ELECTROMETERS

J. T. BOTOMLEY. On Contact Electricity in Common Air, Vacuum and Different

Gases. *Brit. Assoc. Advancement Sci. Rept.*, p. 901 (1885)

A. H. AND K. T. COMPTON. A Sensitive Modification of the Quadrant Electrometer: Its Theory and Use. *Phys. Rev.* **14**, 85 (1919)

H. R. CRANE, C. E. LAURITSEN, A. SOLTAN. Artificial Production of Neutrons. *Phys. Rev.* **45**, 507 (1934)

C. D. GOULD. Subminiature Electrometer Tube. *Electronics* **20.7**, 106 (1947)

K. GRANT. New Electrometer. *J. Sci. Instr.* **9**, 17 (1932)

ROSS GUNN. Principles of a New Portable Electrometer. *Phys. Rev.* **49**, 307 (1932)

W. W. HANSEN. On the General Theory of Electrometer Design. *Rev. Sci. Instr.* **7**, 182 (1936)

L. R. HOFSTAD. The Application of the FP-54 Pliotron to Atomic Disintegration-Studies. *Phys. Rev.* **44**, 201 (1933)

J. CRAMER HUDSON, R. F. COWING. Use of a Lauritsen Electroscope under Adverse Weather Conditions. *Rev. Sci. Instr.* **11**, 347 (1940)

A. W. HULL. Electronic Devices as Aids in Research. *Physics* **2**, 409 (1932)

LORD KELVIN. Contact Electricity of Metals. *Phil. Mag.* **46**, 82 (1898)

C. C. AND THOMAS LAURITSEN. A Simple Quartz Fibre Electrometer. *Rev. Sci. Instr.* **8**, 438 (1937)

F. A. LINDEMANN, A. F. LINDEMANN, T. C. KEELEY. A New Form of Electrometer. *Phil. Mag.* **47**, 577 (1924)

ARTHUR E. LOCKENVITZ. A High Sensitivity String Galvanometer. *Phys. Rev.* **65**, 153A (1944)

F. MÜLLER, K. GEISSLER. Lindemann Electrometer. *Z. Electrochem.* **37**, 857 (1931)

W. W. NICHOLAS. Electrostatic Voltmeter. *J. Research Nat. Bur. Standards* **8**, 111 (1932)

H. PALEVSKY, R. K. SWANK, R. GRENCHIK. Design of Dynamic Condenser Electrometers. *Rev. Sci. Instr.* **18**, 298 (1947)

G. L. PEARSON. Fluctuation Noise in Vacuum Tubes. *Physics* **5**, 233 (1934)

W. F. G. SWANN. The Theory of the Single Fiber Electroscope and a New Design of the Instrument. *J. Optical Soc. Am.* **11**, 375 (1925)

TH. WULF. A Single Fiber Electrometer. *Physik Z.* **15**, 250 (1914)

TH. WULF. A New Electrometer for Static Charges. *Physik Z.* **10**, 251 (1909)

JOHN ZELENY. A Lecture Electroscope for Radio-Activity and Other Ionization Experiments. *Phys. Rev.* **32** (1st series), 581 (1911)

OTHER COUNTING METHODS

Secondary Emission Electron Multipliers

JAMES S. ALLEN. The Detection of Single Positive Ions, Electrons and Photons by a Secondary Electron Multiplier. *Phys. Rev.* **55**, 966 (1939)

JAMES S. ALLEN. The Measurement of the Intensity of X-Rays of Wave-Length 0.14A to 1.0A by Means of an Electron Multiplier Tube. *Phys. Rev.* **59**, 110A (1941)

JAMES S. ALLEN. The X-Ray Photon Efficiency of a Multiplier Tube. *Rev. Sci. Instr.* **12**, 484 (1941)

Z. BAY. Electron-Multiplier as an Electron-Counting Device. *Nature* **141**, 284, 1011 (1938)

Z. BAY. Electron-Multiplier as an Electron-Counting Device. *Rev. Sci. Instr.* **12**, 127 (1941)

M. BLAU, B. DREYFUS. The Multiplier Photo-Tube in Radio-active Measurements. *Rev. Sci. Instr.* **16**, 245 (1945)

D. CHARLES. Electron Multipliers with Magnetic Focusing. *Compt. rend.* **216**, 16 556 (1943)

J. W. COLTMAN, FITZ-HUGH MARSHALL. Some Characteristics of the Photo-Multiplier Radiation Detector. *Phys. Rev.* **72**, A528 (1947)

G. H. DIEKE, H. M. CROSSWHITE. Direct Intensity Measurements of Spectrum Lines with Photo-Multiplier Tubes. *J. Optical Soc. Am.* **35**, 471 (1945)

RALPH W. ENGSTROM. Multiplier Photo-Tube Characteristics: Application to Low Light Levels. *J. Optical Soc. Am.* **37**, 420 (1947)

C. C. LARSON, H. SALINGER. Photo-Cell Multiplier Tubes. *Rev. Sci. Instr.* **11**, 226 (1940)

FITZ-HUGH MARSHALL, J. W. COLTMAN. The Photo-Multiplier Radiation Detector. *Phys. Rev.* **72**, 528A (1947)

FITZ-HUGH MARSHALL, J. W. COLTMAN, L. P. HUNTER. Photo-Multiplier X-Ray Detector. *Rev. Sci. Instr.* **18**, 504 (1947)

L. MALTER. The Behavior of Electrostatic Electron Multipliers as a Function of Frequency. *Proc. Inst. Radio Engrs.* **29**, 587 (1941)

R. D. SARD. Noise Spectrum of 931 Photo-Multiplier Tube. *J. Applied Phys.* **17**, 768 (1946)

W. S. SHOCKLEY, J. R. PIERCE. A Theory of Noise for Electron Multipliers. *Proc. Inst. Radio Eng.* **26**, 321 (1938)

V. K. ZWORYKIN, J. A. RAJCHMAN. The Electrostatic Electron Multiplier. *Proc. Inst. Radio Eng.* **27**, 558 (1939)

V. K. ZWORYKIN, J. E. RUEDY, E. W. PIKE. Silver-Magnesium Alloy as a Secondary Electron Emitting Material. *J. Applied Phys.* **12**, 696 (1941)

V. K. Zworykin, G. A. Morton, L. Malter. The Secondary Emission Multiplier: A New Electronic Device. *Proc. Inst. Radio Eng.* **24**, 351 (1936)

Cerenkov Radiation

P. A. Cerenkov. Visible Radiation Produced by Electrons Moving in a Medium with Velocities Exceeding that of Light. *Compt. rend. acad. sci. U.R.S.S.* **8**, 451 (1934); **12**, 413 (1936); **14**, 102, 105 (1937); *Phys. Rev.* **52**, 378 (1937)

George B. Collins, Victor G. Reiling. Cerenkov Radiation. *Phys. Rev.* **54**, 499 (1938)

R. T. Cox. Momentum and Energy of Photon and Electron in the Cerenkov Radiation. *Phys. Rev.* **66**, 106 (1944)

R. H. Dieke. Cerenkov Radiation Counter. *Phys. Rev.* **71**, 737 (1947)

I. Frank, I. Tamm. Coherent Visible Radiation from Fast Electrons Passing through Matter. *Compt. rend. acad. sci. U.R.S.S.* **14**, 9, 109 (1937)

I. A. Getting. A Proposed Detector for High Energy Electrons and Mesons. *Phys. Rev.* **71**, 123L (1947)

W. H. Furry. Discussion of a Possible Method for Measuring the Masses of Cosmic-Ray Mesotrons. *Phys. Rev.* **72**, 171A (1947)

H. O. Wyckoff, J. E. Henderson. The Spacial Asymmetry of Cerenkov Radiation as a Function of Electron Energy. *Phys. Rev.* **58**, 187A (1940), **64**, 1 (1943)

Photographic Registration of Single Particles

W. H. Bragg, J. P. V. Madsen. An Experimental Investigation of the Nature of Gamma-Rays. *Phil. Mag.* **16**, 918 (1908)

E. Broda. Radiation Track Counts in Photographic Plates. *J. Sci. Instr.* **24**, 136 (1947)

G. R. Evans, C. G. Griffiths. Slow Cosmic-Ray Mesons at Sea Level. *Nature* **159**, 879 (1947)

Hugh T. Richards, Emmett Hudspeth. Photographic Plate Spectrum of D-D Neutrons. *Phys. Rev.* **58**, 382L (1940)

H. T. Richards. A Photographic Plate Spectum of the Neutrons from the Disintegration of Lithium by Deuterons. *Phys. Rev.* **59**, 796 (1941)

E. Rutherford, H. Geiger. Photographic Registration of Alpha-Particles. *Phil. Mag.* **24**, 618 (1912)

Miscellaneous

W. Y. Chang, S. Rosenblum. A Simple Counting System for Alpha-Ray Spectra and the Energy Distribution of Polonium Alpha-Particles. *Phys. Rev.* **67**, 222 (1945)

H. Greinacher. Acoustic Observation and Galvanometric Registration of Elementary Rays and Single Ions. *Z. Physik* **23**, 371 (1924)

H. Greinacher. Mains-Operated Spark-Counter. *Z. tech. Physik* **19**, 132 (1938)

S. H. Neddermeyer, E. J. Althaus, W. Allison, E. R. Schatz. Measurement of Ultra-Short Intervals. *Rev. Sci. Instr.* **18**, 488 (1947)

S. Rosenblum, W. Y. Chang. A Simple Counting System for Alpha-Ray Spectra. *Phys. Rev.* **67**, 58A (1945)

Robert N. Varney. The Sensitivity of the Balanced Space-Charge Positive Ion Detector. *Phys. Rev.* **53**, 732 (1938)

D. E. Wooldridge, A. J. Ahearn, J. A. Burton. Conductivity Pulses Induced in Diamond by Alpha-Particles. *Phys. Rev.* **71**, 913L (1947)

STATISTICS

H. Bateman. On the Probability Distribution of Alpha-Particles. *Phil. Mag.* **20**, 704 (1910)

Karl K. Darrow. Statistical Theories of Matter, Radiation and Electricity. *Revs. Mod. Phys.* **1**, 90 (1929)

Karl K. Darrow. Memorial to the Classical Statistics. *Bell. Sys. Tech. J.* **22**, 108 (1943)

Willy Feller. On the Time Distribution of So-Called Random Events. *Phys. Rev.* **57**, 906 (1940)

E. J. Gumbel. The Limiting Form of Poisson Distribution. *Phys. Rev.* **60**, 689L (1941)

N. Hale. On the Distribution of Counts in a Counting Apparatus. *Arkiv. Mat. Astron. Fysik* **33B** (No. 3) Paper 8 (1947)

J. Von Juilfs. Calculation of Accidental Multiple Coincidences. *Physik Z.* **40**, 697 (1939)

Edwin C. Kemble. Is the Frequency Theory of Probability Adequate for All Scientific Purposes? *Am. J. Phys.* **10**, 6 (1942)

Eric Rogers. Probable Error for Poisson Distributions. *Phys. Rev.* **57**, 735 (1940)

Arthur E. Ruark. Note on Events Distributed According to Chance. *Phys. Rev.* **65**, 287L (1944)

D. J. Strink. Derivations of Poisson's Law. *J. Math. Phys.* **9**, 151 (1929-30)

NUCLEONIC EVENTS

UNIV. OF CHICAGO REPLACED AS CLINTON CONTRACTOR

Reversing itself for the second time on its plans for the Clinton National Laboratory at Oak Ridge, Tennessee, the Atomic Energy Commission has requested the University of Chicago to withdraw from its contract for operation of the laboratory (which it was to have assumed January 1) and has made arrangements for the Carbide and Carbon Chemicals Corporation to be the new operator. Carbide and Carbon already operates the diffusion and electromagnetic plants at Oak Ridge for production of U-235.

Last fall, the Monsanto Chemical Company decided to quit its operation of Clinton as a result of policy disagreements with AEC and also, apparently, because of rather strained relations between the company and laboratory personnel. At that time, the plan was for the University of Chicago to assume operation of Clinton.

The decision to have Carbide and Carbon take over instead is the result of a change in operating policy for Clinton. This laboratory is now to concentrate on applied research in chemistry and chemical engineering. Reactor development previously planned for Clinton and for the Argonne National Laboratory at Chicago will be consolidated at Argonne under the direction of Walter H. Zinn.

Carbide and Carbon has agreed to assume operation of Clinton not later than March 1. Monsanto will continue as operator until that date.

The shift in program at Clinton culminates a trend which has been in evidence ever since AEC succeeded the Manhattan District. During its postwar period of operation, the Army had set up Clinton as a center for pile work. Work was started at that time on a demonstration pile, the so-called Daniels pile, for production of electric power. Under the AEC, Argonne has been built up as the center for pile research; power production problems have been assigned to the General Electric Company's Knolls Laboratory; and work on the Daniels pile has been dropped. Clinton's new concentration on chemical problems completes the process

—Robert Colborn
McGraw-Hill Washington Bureau

OFFICE OF NAVAL RESEARCH REVIEWS 1947 PROGRESS

The Office of Naval Research has listed the following important results from its nuclear physics program for the past fiscal year:

Measurement of the magnetic moment and spin of the third simplest nucleus, H^1, has been accomplished. This, it is said, will be of great use in calculation of nuclear forces.

The rate of absorption of mesons by matter has been determined—information regarded as vital in the interpretation of nuclear forces.

Preliminary evidence for the existence of the neutrino has been discovered. This postulated fundamental particle, ONR states, is believed to be capable of penetrating great distances of solid matter.

Important efforts in nuclear science, especially fundamental basic research in the universities during the fiscal year 1947, are summed up by ONR as follows:

Particle Accelerator Construction. Partial support is being given for five frequency-modulated cyclotrons for heavy particle acceleration, at Columbia University, Harvard University, University of Rochester, Carnegie Institute of Technology, and the University of Chicago; for five synchrotrons for electron acceleration, at Massachusetts Institute of Technology, Purdue University, Cornell University, and two different types at the General Electric Company; and for four different types of linear electron accelerators being developed at the Franklin Institute, Purdue University, Stanford University, and Yale University.

Nuclear Energy Levels and Forces. The routine investigations of nuclear energy levels and forces, ONR believes, will lead to many useful applications. ONR has placed contracts with five institutions for the construction of precision beta- and gamma-ray spectrographs for studying the energy levels in radioactive disintegrations.

A second group of projects on energy levels, according to ONR, includes photo-disintegration work, slow neutron absorption measurement, and study of reactions with separated isotopes; also short-lived isotopes, very short-lived metastable levels, cross sections and angular distribution for various light-element collisions, and cross sections for gamma-ray processes.

Medical, Biological, and Chemical Radiation Studies. Scientists at the California Institute of Technology, working under ONR contract, have announced an important achievement during the past year in the use of radioactive isotopes in the *in vitro* vital tissue synthesis of urea. This will facilitate protein-synthesis study.

Studies are also under way, sponsored by ONR, on the effects of 3-Mev X-ray beams on living tissue, and on the bactericidal effects of such high-energy radiation.

Miscellaneous Instrumentation. Numerous instruments, each designed for a specific function, will be developed for measuring the properties of high-energy particles. ONR is supporting research for the design and construction of such apparatus as high-pressure cloud chambers, beta-ray spectrometers, mass spectrometers, ion-velocity selectors, improved Geiger counters, crystal counters and electron multipliers, photographic plates for recording neutrons, and coincidence circuits having a resolving time around one hundred-millionth of a second.

—Gladys T. Montgomery
McGraw-Hill Washington Bureau

PAULING PRESIDENT-ELECT OF ACS

Linus C. Pauling, chairman of the division of chemistry and chemical engineering of the California Institute of Technology, has been elected president of the American Chemical Society for 1949. Dr. Pauling took office as president-elect on January 1, when Charles A. Thomas, executive vice president and technical director of the Monsanto Chemical Co., St. Louis, Mo., became president of the society. Dr. Thomas was recently selected to receive the 1948 gold medal of The American Institute of Chemists.

MEETINGS

American Institute of Mining and Metallurgical Engineers—Pennsylvania Hotel, New York, N. Y., Feb. 15–19

Electrochemical Society—Columbus, O., April 14–17

American Chemical Society—113th Annual Meeting, Chicago, Ill., April 19–23

American Association of Petroleum Geologists—Denver, Col., April 26–29

American Society of Medical Technologists—St. Paul, Minn., June 7–9

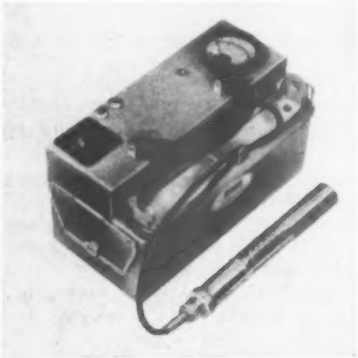
Pittsburgh International Conference on Surface Reactions—Mellon Institute of Industrial Research, Pittsburgh, Pa., June 7–11

Institute of Radio Engineers—6th Annual Electron Tube Conference, Cornell University, Ithaca, N. Y., June 28, 29

PRODUCTS and MATERIALS

PORTABLE G-M COUNTER

Beckman Instruments, National Technical Laboratories, South Pasadena, Calif. The model MX-5 portable (weighs 9 pounds) G-M counter is used to detect and measure X, beta, and gamma radiations. The battery-operated meter has a self-quenching G-M



tube mounted on a three-foot probe and has three operating ranges, 20, 2 and 0.2 mr per hr. The circuit is claimed to eliminate zero control, to feature rapid meter response and to extend the battery life to 400 hours.

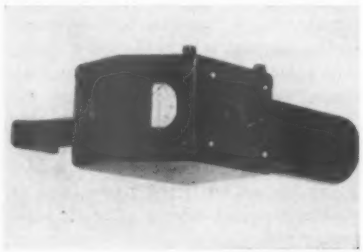
MASS SPECTROMETER

Process and Instruments, 60 Greenpoint Ave., Brooklyn 22, N. Y. The model M60 mass spectrometer has been especially designed for the analysis of isotopic and complex gas mixtures of interest in the biochemical field; thus, it possesses its optimum resolution in the "biochemical" range up to mass 100. The variable magnetic field permits larger masses to be focussed with lower

resolution. The M60 is a 60-degree-type instrument, is completely operated from a 115 volt, 60 cps line. Mass scanning is performed by manually varying the accelerating voltage with a voltage divider. Ion beam intensity for each mass is measured with a potentiometer by a nullpoint method.

"CUTIE PIE"

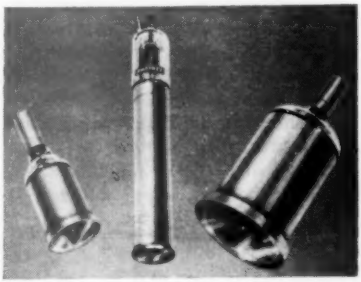
Tracerlab, Inc., 55 Oliver St., Boston, Mass. This portable beta and gamma radiation meter is a redesigned version of the recently declassified "Cutie Pie." It is an ionization-chamber-type meter featuring small dimensions (4 in. × 6 in. × 5 in.) and weight (5 lbs.), especially designed for use in so-called "hot" laboratories handling levels of radio-



activity of the order of millicuries or hundreds of rutherfords. Three full-scale ranges of approximately 100, 1,000, and 10,000 mr per hr are provided. It is claimed that a thin window at the end of the ionization chamber permits entry of beta particles of less than 0.1 Mev; also a permanently mounted shield can be used to eliminate beta radiation.

GEIGER TUBES

Amperex Electronic Corp., 25 Washington St., Brooklyn 1, N. Y. The Amperex G-M tubes have been redesigned for standardized production. The first series to be released includes counter



tubes for beta, gamma and X-rays. Brochures describing these tubes are available from the manufacturer.

POCKET DOSIMETER

Instrument Development Laboratories, 229 W. Erie St., Chicago 10, Ill. The model 3360 pocket dosimeter provides an indication of the amount of radioactivity to which the carrier of the instrument has been exposed. It can be read by looking through the cupped eyepiece toward a light source. The aluminum housing is claimed not to distort appreciably the free air reading. A molded, flake-proof, conducting plastic cathode surrounds the ion chamber, and the anode comprises a sensitive fused quartz fiber, 5 microns in thickness.

HIGH-VOLTAGE POWER SUPPLY

Atomic Instrument Co., 160 N. Charles St., Boston 14, Mass. This 500–2000 volt power supply delivers 500 microamperes at 2,000 volts, and 1 ma. for voltages up to 1,500. It is claimed that regulation is maintained within 1% for ±10% line voltage variations and load variations up to maximum rated load. At any voltage setting, the drift is said to be within 0.1%. Six miniature tubes are used, and the complete unit

is housed in a cabinet measuring 7½ in. × 12 in. × 5¼ in. and weighing 9½ lbs.

GEIGER TUBE

In a description of the type 410-A Geiger tube, manufactured by the **Cyclotron Specialties Co., Moraga, Calif.**, in the November issue of NUCLEONICS, the statement that the "tube, when shielded, reportedly has a background of approximately 1% in the G-M region" should have read "background of approximately one count per sec."

ABSTRACTS

CHEMICAL PUBLICATIONS

Ion-exchange as a separations method. I. The separation of fission-produced radioisotopes, including individual rare earths, by complexing elution from Amberlite resin, E. R. Tompkins, J. X. Khym, W. E. Cohn (Clinton National Laboratory, Oak Ridge, Tenn.) *J. Am. Chem. Soc.* **69,** 2769–2777 (1947). The major radioactive fission species existing in uranium were separated nearly quantitatively in radiochemically and chemically pure form and without resort to carrier materials by a process of elution from ion-exchange columns with complex-forming reagents. The separations were carried out on microcurie up to curie levels.

For production runs the procedure was as follows: The Zr-Cb mixture was eluted by 0.5% oxalic acid. Y was then eluted with 5% citrate at a pH 2.7, after which the pH was shifted at once or gradually to about 3.0 and the rare earths collected successively until Ce appeared. Sr and Ba were then removed in succession by 5% citrate at a pH 5. Ru and Te may be recovered from the effluent of the original adsorption step. The degree of separation of the individual rare earths depends upon the elution rates and pH's used. Large masses of material may be separated in this way by increasing the mass of resin used so as to keep the same ratio of cation mass to resin mass.

The chemical identification of radioisotopes of neodymium and of element 61, J. A. Marinsky, L. E. Glendenin, C. D. Coryell (Massachusetts Inst. of Technology, Cambridge), *J. Am. Chem. Soc.* **69** 2781–2785 (1947). The results of ion-exchange studies using Amberlite IR-1 show clearly that the order of elution of the rare earths is the reverse of that of atomic number, with yttrium being eluted in the region of gadolinium. The atomic number of an unknown radioisotope can be determined from the elution curves of a group of successive rare earths, and useful separations of rare earths can be achieved. The procedure involves adsorption of the rare earth ions on a column of the resin from a dilute acid solution, followed by elution with a 5% ammonium citrate solution adjusted to a pH of about 3. A mixture of fission products containing activities of praseodymium, neodymium and element 61 which was subjected to such an ion-exchange separation, resulted in the positive chemical identification, for the first time, of the 47-hr and 3.7-yr isotopes of element 61. Radiochemical studies were carried out on 11-d Nd^{147}, 1.7-hr Nd^{149}, 3.7-yr 61^{147} and 47-hr 61^{149}, and their decay characteristics, genetic relationships, fission yields, and mass assignments were established.

Techniques in the use of carbon-14, W. G. Dauben, J. C. Reid, P. E. Yankwich (Univ. of California, Berkeley), *Ind. Eng. Chem., Anal. Ed.* **19,** 828–832 (1947). A method is given for incorporating $C^{14}O_2$ into organic compounds by carbonation of the Grignard reagent. Pure acids were obtained in yields of 80 to 95% based on the carbon dioxide. A modified Pregl-Grant microanalysis setup was used to convert the isotopic organic compound into barium carbonate, and at the same time to determine the carbon content. The samples for counting consisted of thin layers of barium carbonate deposited on aluminum discs. A special thin mica window bell-type G-M tube containing a grid to support the mica was used. Detailed descriptions are given throughout.

Properties and measurement of carbon-14, A. F. Reid, A. S. Weil, J. R. Dunning (Columbia Univ., New York), *Ind. Eng. Chem., Anal. Ed.* **19,** 824–827 (1947). The mean absorption coefficients in aluminum and cellophane of C^{14} were found to be 247 sq cm per gram and 282 sq cm per gram respectively. Techniques are given for the measurement of C^{14} samples outside a thin mica-window Geiger counter (window thickness, 2 to 6 mg per sq cm). Corrections for resolving time, background, efficiency changes with time, geometry, and absorption are discussed in detail. The effect of statistical accuracy of counting on the precision of measurements is given. Illustrative procedures for C^{14} measurements are outlined for three cases: (1) when only relative measurements are desired and plenty of sample material is available; (2) when only relative measurements are desired and sample material is limited; (3) where absolute measurements of sample activity are wanted.

Long-lived radioactivities induced in rhodium and tungsten by neutrons, O. Minakawa, *Sci. Papers Inst. Phys. Chem. Research* (Tokyo) **42,** 109–114 (1944). W and Rh were bombarded with fast and slow neutrons, and the activities of the products studied. The following mechanism is suggested: $Rh^{103}(n,2n)Rh^{102}$ (102 days, e^-, e^+); $W^{184}(n,\gamma)W^{185}$; $W^{186}(n,2n)W^{185}$ (76 days, e^-).

Theory of mesotrons. I, S. Tomonaga, *Sci. Papers Inst. Phys. Chem. Research* (Tokyo) **39,** 247–266 (1941). The Hartree approximation is used to treat a theory of mesotrons.

Theory of mesotrons. II, T. Miyajima, S. Tomonaga, *Sci. Papers Inst. Phys. Chem. Research* (Tokyo) **40,** 21–67 (1942). Mesotron formation is discussed by comparison with the results of chemical treatment.

Magnetic scattering of slow neutrons in a ferromagnetic crystal, T. Muto, M. Nogami, *Sci. Papers Inst. Phys. Chem. Research* (Tokyo) **40,** 333–350 (1943). Inelastic, magnetic scattering of neutrons in the ferromagnetic crystal is treated, and the effective cross section is worked out.

Inelastic scattering of fast neutrons accompanied by the excitation of the nuclear surface vibrations of the liquid-drop model, T. Muto, *Sci. Papers Inst. Phys. Chem. Research* (Tokyo) **40,** 351–363 (1943). Using the liquid-drop model, the effective scattering cross section of atomic nuclei for fast neutrons was calculated. The surface structure of the atomic nucleus appears to be an essential factor in nuclear reactions.

Pair production by γ-rays from fluorine bombarded with protons. III, M. Hatoyama, J. Yuhara, T. Malyama, *Sci. Papers Inst. Phys. Chem. Research* (Tokyo) **39,** 1–7 (1941). Gamma-rays of 6.25 ± 0.05 Mev produced pairs in the gas of the cloud chamber. The probability of distribution of energy between positive and negative electrons was studied.

Decay of negative mesons in matter, H. Fröhlich (Univ. of Bristol, Royal Fort, Bristol), *Nature* **160,** 255 (1947). If an assembly of electronic oscillators is used as a model, the slowing-down time of mesons in solids is many orders of magnitude smaller than the times calculated using the Fermi gas of free electrons as a model. Therefore, the drastic change in the present conception of the interaction of mesons with atomic nuclei suggested by Fermi, Teller and Weisskopf does not appear to be warranted.

Definition of nuclear quadrupole moments, E. H. Rhoderick (Cambridge Univ.), *Nature* **160,** 255–256 (1947). There is some confusion in the literature

over the definition of the quadrupole moment of the nucleus. In interpreting reported values of the quadrupole moment, it should be realized that the experimentally determined quantity is $Q = \int \rho(3z^2 - r^2)d\tau$ and not Q/Z where Z is the total number of protons.

Effect of gamma- and X-rays on dilute aqueous solutions of acrylonitrile, F. S. Dainton (Cambridge Univ.), *Nature* **160,** 268–269 (1947). Using a γ-ray source, the polymerization of acrylonitrile was found to be second order with respect to the monomer concentration, and independent of the source strength. The ion-pair yield was at least 10^4 for the 600 millicurie source at the highest concentration used, and varied inversely with the source strength. These results are explained by assuming that the radiation acts upon the water molecules to produce free hydrogen atoms and hydroxyl radicals, and that these products initiate the polymerization chain reaction.

"Evaporation" of heavy nuclei, D. H. Perkins (Imperial College of Science and Technology, London), *Nature* **160,** 299–301 (1947). The energy distribution of the particles was determined from the disintegration of fifteen heavy nuclei (silver and bromine) out of a total of some two hundred stars found on 400 sq cm of photographic plate area exposed to cosmic radiation.

A histogram of the energy distribution of a total of eighty star proton tracks was constructed, and compared with the calculated energy spectrum of charged particles evaporating from a highly excited nucleus in thermal equilibrium. The proportion of protons at the low-energy end indicates that the potential barrier height is not a well-defined quantity. The high-energy tail of the spectrum indicates that some of the particles on the nuclear surface leave the nucleus immediately with a very high energy upon being struck by the incident particle. The angular distribution of tracks in a number of stars shows marked deviations from spherical symmetry, which is explicable on the basis that thermal equilibrium is not fully attained.

The one-million volt accelerating equipment of the Cavendish Laboratory, Cambridge, W. E. Eurcham (Cambridge Univ.), *Nature* **160,** 316–318 (1947). A description is given of the performance of, and type of work carried out with, the million-volt apparatus since it was installed ten years ago.

Observations underground of penetrating cosmic ray showers, E. P. George, A. C. Jason (Birkbeck College, Univ. of London), *Nature* **160,** 327–328 (1947). Measurements of penetrating showers and penetrating bursts were made in a laboratory approximately 100 ft under clay. The strong decrease of the intensity of these two events supports the hypothesis that penetrating showers are produced by primary nucleons, and is not in agreement with the assumption that penetrating showers arise from the hard component itself. No production of penetrating particles was recorded in 100 days.

Traveling-wave linear accelerator for electrons, D. W. Fry, R. B. R. S. Harvie, L. B. Mullett, W. Walkinshaw (Telecommunications Research Estab., Great Malvern, Worcs.), *Nature* **160,** 351–353 (1947). Electrons are accelerated continuously by carrying them in tight bunches around a stable position in a traveling radio-frequency wave. A traveling wave (10.0-cm wavelength) is set up in a circular waveguide 40 cm long having an axial electric field. The phase velocity of this wave is adjusted by varying the depth of deep corrugations in the wall of the guide so as to keep it in step with the velocity of the electron as it is

accelerated. The phase velocity is 0.4 c at injection and 0.875 c at exit. The energy spectrum shows a maximum amplitude at an energy of about 540 kev, and the width between half-amplitude points was 65 kev when the applied r-f power was 1 megawatt. Measurements show that a very large proportion of the injected electrons became trapped by the wave and were brought into stable bunches. The present 40 cm length of accelerator is only the first section of one which is designed to operate best at 20 meters.

. . . I. W. RUDERMAN

LIFE SCIENCE PUBLICATIONS

On the mechanism of enzymatic conversion of glucose-1-phosphate to glucose-6-phosphate, M. Schlamowitz, D. Greenberg (Division of Biochemistry, Univ. of California Medical School, Berkeley), *J. Biol. Chem.* **171,** 293–295 (1947). Three possible mechanisms are presented for the enzymatic conversion of glucose-1-phosphate to glucose-6-phosphate. The first was shown to be in doubt by another worker and the second mechanism is shown to be unlikely by the present work. Evidence favoring an intramolecular mechanism for the enzymatic conversion of glucose-1-phosphate to glucose-6-phosphate is presented. A cyclic phosphate diester is proposed as the intermediate in this reaction.

On the nature of plasma iodine, A. Taurog, I. Chaikoff (Division of Physiology, Univ. of California Medical School, Berkeley), *J. Biol. Chem.* **171,** 439–440 (1947). Evidence is presented that the circulating thyroid hormone is thyroxine by showing that approximately 90% of suspected plasma thyroxine behaves like thyroxine in its solubility properties.

Incorporation of labeled glycine into the protein of tissue homogenates, F. Friedberg, T. Winnick, D. Greenberg (Division of Biochemistry, Univ. of California Medical School, Berkeley), *J. Biol. Chem.* **171,** 441–442 (1947). Incubated liver and spleen homogenates took up 0.1% of glycine containing C^{14} employed in the media. Homogenates boiled prior to incubation showed almost no activity. Intestine homogenates showed no significant uptake.

The metabolism of acetic acid in animal tissues, K. Bloch (Dept. of Biochemistry and Inst. of Radiobiology and Biophysics, Univ. of Chicago), *Physiol. Revs.* **27,** 574–620 (1947). An extensive review discussing the metabolism of acetic acid, its occurrence in animal tissues, the acetylation reaction, metabolism of fatty acids, formation of acetate from pyruvate, the use of acetic acid for synthetic reactions, formation of protoporphyrin, uric acid, the biologically active form of acetic acid and related material (with 191 references).

Isotopic composition of acetylmethylcarbinol formed by yeast juice, N. Gross, C. Werkman (Dept. of Bacteriology, Iowa State College, Ames), *Arch. Biochem.* **15,** 125–131 (1947). Acetaldehyde containing C^{13} in both carbon atoms was reacted with pyruvic acid in the presence of active dried yeast juice to form heavy carbon acetylmethylcarbinol. The compound was degraded and the C^{13} content of each of the carbon atoms obtained. It was found that the heavy carbon of the acetaldehyde had gone into all carbons of the acetylmethylcarbinol. It is believed likely that acetylmethylcarbinol is formed by condensation of pyruvic acid and acetaldehyde. Distribution of C^{13} among the four carbon atoms was not uniform. This is believed to be due to a partial conversion of acetylmethylcarbinol to a symmetrical compound. Other methods of formation are not excluded. It is not likely that

the two molecules of labeled acetaldehyde condense to form the carbinol because this would result in uniform distribution of the acetaldehyde heavy carbon which was not the case.

Distribution of a large dose of thyroxine labeled with radioiodine in the organs and tissues of the rat, J. Gross, C. Leblond (Dept. of Anatomy, McGill University, Montreal, Canada), *J. Biol. Chem.* **171,** 309–320 (1947). DL-thyroxine containing radioactive iodine was prepared and administered to rats in large amounts. The distribution of this compound in a great number of organs and tissues at the end of 2 and 24 hours was studied. At the end of the 2-hour period more than 50% of the radioactivity was detected in the gastrointestinal tract, pancreas, liver and stomach. Except for the stomach, the activity was due mainly to thyroxine-bound iodine. Only 2% of the radioactivity was detectable in the circulating plasma. Significant concentrations were also found in the kidneys, lungs, adrenals, lymphatics and ovaries.

At the end of the 24-hour period about 80% of the radioactivity was found in the feces largely in the form of unchanged thyroxine. About 11% of the dose was detected as inorganic iodide in the urine. The radioactivity concentrations of the pancreas, liver, adrenals, kidneys, ovaries and lymphatic tissue remained fairly constant.

The liver and bile were mostly responsible for the entry of thyroxine into the gastrointestinal tract. In about 24 hours the great bulk of the administered thyroxine is eliminated from the body. About 25% of the thyroxine is destroyed in 2 hours and about 50% at the end of 24 hours.

The fixation of carbon dioxide in a plant tricarboxylic acid system, B. Vennesland, J. Ceithaml, M. Gollub (Dept. of Biochemistry, Univ. of Chicago), *J. Biol. Chem.* **171,** 445–446 (1947). Evidence is presented that in plants tricarboxylic acids may be formed by a mechanism involving the addition of carbon dioxide to α-ketoglutarate, in a manner similar to that demonstrated in animal tissues.

The mechanism of iodine concentration by the thyroid gland: its nonorganic iodine-binding capacity in the normal and propylthiouracil-treated rat, A. Taurog, I. Chaikoff, D. Feller (Division of Physiology, Univ. of California Medical School, Berkeley), *J. Biol. Chem.* **171,** 189–201 (1947). Rat thyroid glands were made goitrous by feeding of propylthiouracil. These glands were able to concentrate injected iodine although they could not form organic iodine compounds. Their capacity to fix injected iodine as nonorganic iodine was much greater than that of normal glands.

Similar curves were obtained for the changes in inorganic iodine concentration in thyroid tissue and plasma after injection of iodide for goitrous rats. A maximum concentration was obtained in one-half hour and the iodide concentration fell rapidly thereafter. The ratio of iodide concentration in goitrous thyroid and in plasma was about 250 to 1 for all periods observed. This suggests that the iodide trapping by the goitrous thyroid is limited by plasma iodide concentration.

The iodine taken by the goitrous thyroid gland is held to be all in the form of inorganic iodide and not stably bound to protein although the possibility of a labile protein-iodine bonding is not ruled out. The thyroids of rats treated with propylthiouracil for 12 hours were normal in size but were not able to synthesize organic iodine compounds. However, they showed the same initial rate of iodine fixation of injected iodide as did control glands. This suggests that in the normal thyroid an iodine fixing mechanism

exists which is independent of the conversion of iodine to diiodotyrosine and thyroxine. The nature of the linkage that binds the iodide in the thyroid gland is discussed.

The metabolism of isotopic lactic acid and alanine in the phlorhizinized animal, S. Gurin, A. Delluva, D. Wilson (Dept. of Physiological Chem., School of Medicine, Univ. of Pennsylvania), *J. Biol. Chem.* **171,** 101–110 (1947). DL-alanine containing C^{13} in the carboxyl position and lactic acid containing C^{14} were prepared by previously described methods. These compounds were fed separately to phlorhizinized rats. Urines were collected and urinary glucose isolated in each case. Of the administered alanine, 60 to 70% appeared to be converted to "extra" urinary glucose, although only a very small percentage of the administered isotope was recovered therein.

About half of the administered lactic acid isotope was converted to urinary ketone bodies and fat. Approximately 28% of the C^{14} was accounted for by urinary glucose formation. It is believed that lactate, unlike alanine, is rapidly converted to glucose. It is also believed that alanine is incorporated into tissue protein to a large extent and that only a small portion enters the circulating carbohydrate pool through oxidative deamination. All the carbons of the excreted acetoacetate and β-hydroxybutyrate contained radioactivity. It is believed that, in the phlorhizinized rat, lactate may be converted directly to ketone bodies to a considerable extent without intermediate formation of fat.

Incorporation of C^{14} from carboxyl-labeled DL-alanine into the proteins of liver slices, I. Frantz, Jr., R. Loftfield, W. Miller (Medical Laboratories, Collis P. Huntington Memorial Hospital of Harvard Univ., Massachusetts General Hospital, and Dept. of Physics, Massachusetts Inst. of Technology), *Science* **106,** 544–545 (1947). Carboxyl-labeled DL-alanine was prepared by the Strecker synthesis modified for the use of small amounts of radioactive hydrogen cyanide. Rat liver slices were incubated in Warburg flasks with the labeled alanine, buffer, and KOH in the center well. After incubation for varying periods of time, the tissue protein was isolated, hydrolyzed, and fractionated. The amino acids were decarboxylated using the ninhydrin reaction and the CO_2 analyzed for activity.

The incorporation of activity into the trichloric acid precipitable protein was slow and dependent upon the utilization of oxygen by the tissue. Preliminary experiments indicated that a large part of the activity is accounted for by the alanine fraction. No activity was detected in the glycine fraction. It is not believed likely that the presence of activity in the protein is due to transcarboxylation.

Conversion of isotopically enriched CO_2 to CO, R. Bernstein, T. Taylor (Chemistry Dept., Columbia Univ.), *Science* **106,** 498–499 (1947). This method makes use of the isotopic exchange reaction between C^{13} carbon monoxide and normal carbon dioxide to form labeled carbon dioxide and normal carbon monoxide in conjunction with thermal diffusion which produces countercurrent flow of CO_2 and CO. $C^{13}O_2$ produced by isotopic exchange at a hot wire is transported toward the bottom of the column where it is converted to labeled carbon monoxide to maintain total reflux. Zinc powder supported on asbestos fiber was used for the conversion. Various flow rates and temperatures were studied. Conversion was almost complete in 5 to 6 hours at 425° and 485° C.

Incorporation of C^{14}-labeled glycine into intestinal tissue and its inhibition by azide, T. Winnick, F. Friedberg, D.

Greenberg (Medical School, Division of Biochemistry, Univ. of California, Berkeley), *Arch. Biochem.* **15,** 160–161 (1947). Heat, the mechanical disintegration of cells, and sodium azide were found to be effective inhibitors in the uptake of glycine by intestinal tissue. Untreated tissue cut into squares and incubated with buffer, radioglycine in a 95% oxygen, carbon dioxide atmosphere for 90 minutes resulted in some incorporation. The inhibitory action of sodium azide, it is suggested, is due to some direct action on the enzymes which promote peptide bond formation.

The transmission of radiostrontium and plutonium from mother to offspring in laboratory animals, M. Finkel (Biology Division of Metallurgical Laboratory and Argonne National Laboratory, Chicago, Ill.), *Physiol. Zoöl.* **20,** 405–421 (1947). Mice injected with radioactive Sr^{89} before conception had fewer litters than the controls. Injection of radioactive strontium or of plutonium increased the number of stillborn litters and the number of stillborn young in the litters that lived. Pregnant mice treated with either Sr^{89} or Pu lived longer than similarly treated nonpregnant female mice. Treated pregnant mice also suffered less radiation damage.

After delivery, total Sr retained by mothers was the same as that of virgin females at similar time intervals after injection. The proportion of Sr or Pu transmitted to the young at birth varied with the injection-delivery period. When the injection-delivery period was within 4 days, the Sr activity per gram body weight of the newborn mouse exceeded that of the mother. The initial excretion of radioactive Sr by very young mice was considerably lower than that of adult mice.

Long bone malformation, anemia, retardation of growth, and osteogenic sarcoma occurred before weaning in animals that were treated in utero with Sr^{89}. No pathologic conditions developed in the animals treated with Pu. No difference in sensitivity to Pu or Sr^{89} was found in young and adult tissues.

. . . BERNARD KANNER

PHYSICAL PUBLICATIONS

Electrostatic analyzer for 1.5 Mev protons, W. Fowler, C. Lauritsen, T. Lauritsen (California Inst. of Technology, Pasadena) *Rev. Sci. Instr.* **18,** 818–820 (1947). To obtain bombarding particles homogeneous in energy to a few hundred volts in a million, external stabilization is necessary for long runs. Both electric and magnetic fields are necessary to separate out deuterons of half-energy as well as protons of different energy. The system described consists of a weak magnetic field to give rough separation of the beam into component masses, together with an electric field whose force is perpendicular to that of the electric field. It separates out protons of different energies, and covers 90°. Collimating slits take out the undesirable protons. The generator voltage is controlled by the analyzing system, and this allows an energy spread of 300 volts in a million, with a beam current of a microampere.

Scale expander for mass-spectrometer recorder, D. Grove, J. Hipple (Westinghouse Research Laboratories, East Pittsburgh, Pa.) *Rev. Sci. Instr.* **18,** 837–841 (1947). A method is described which gives automatic recording on a mass spectrometer with a scale automatically chosen by an automatic shunt selector so as to give wide range of peak heights without loss of accuracy.

Two new methods for detecting nuclear radio-frequency resonance absorption, A. Roberts (State Univ. of Iowa, Iowa City), *Rev. Sci. Instr.*, **18,** 845–848 (1947). The hitherto used methods

involve the unbalancing of a bridge or production of a voltage in a secondary coil by the nuclear absorption of a radio-frequency field. The first new method involves placing inductance of a super-regenerative circuit in a magnetic field and placing it in the sample. At resonance, the absorption is equivalent to an increased resistance which reduces the integrated pulse energy. (A quench circuit is used.) The other method is an autodyne, or weakly oscillating detector, which provides a signal beating against an incoming signal to give a heterodyne audio signal. This method uses absorption rather than induction.

Measurement of transport and inelastic scattering cross sections for fast neutrons. I. Method. H. Barschall, J. Manley, V. Weisskopf (Los Alamos Scientific Laboratory, N. M.), *Phys. Rev.* **72,** 875–880 (1947). A theory is given to correspond to the method for determining the values of the inelastic cross section (scattered neutrons having energy different from incident ones) and the transport cross section, which differs from the total cross section in having neutrons scattered through angles θ weighted by the factor $1 - \cos \theta$. A threshold detector was used which counted all neutrons above a certain energy with approximately the same efficiency. Calculations were made for measurements using poor geometry and back-scattering.

Measurement of transport and inelastic scattering cross sections for fast neutrons. II. Experimental results. H. Barschall, M. Battat, *et al.* (Los Alamos Scientific Laboratory, N. M.), *Phys. Rev.* **72,** 881–888 (1947). Measurements discussed in the previous article were carried out with neutrons of various energies from 0.2 to 3 Mev on various elements from C to Pb. Values of inelastic and transport cross sections are given.

Thermal neutron activation cross sections, L. Seren, H. Friedlander, S. Turkel (Argonne National Laboratory, Chicago), *Phys. Rev.* **72,** 888–901 (1947). The activation method consists of measuring the number of beta rays given off by unstable isotopes resulting from thermal neutron capture. Knowing the thermal neutron flux and number of atoms doing the capturing, the capture cross section can be calculated. In some cases there are daughter activities, which allow a check on the results. Results are given for 65 elements, along with the properties of resulting radioisotopes.

Cloud-chamber energy measurements of photo-neutron sources, D. Hughes, C. Eggler (Argonne National Laboratory, Chicago), *Phys. Rev.* **72,** 902–907 (1947). Measurement of the energy distribution of neutrons produced in photo-neutron reactions were made by means of the range distribution of recoil protons in a hydrogen-filled cloud chamber. Background of beta tracks produced by the intense gamma activity of the source was reduced by adjusting the expansion ratio and surrounding the photo-neutron source with lead (which does not change neutron energy because at these energies the collisions are elastic). The results show that the neutron energy is spread out by slowing down in the source.

Scattering of neutrons in polycrystals, R. Finkelstein (Argonne National Laboratory, Chicago), *Phys. Rev.* **72,** 907–913 (1947). The calculations for the scattering of neutrons in a crystal of the Einstein model are given. They are found to approximate Fermi's results for amorphous scatterers, except for higher energies where the struck nucleus can be ejected from the lattice.

Calculations in the liquid-drop model of fission, S. Frankel, N. Metropolis (Institute for Nuclear Studies, Chi-

cago), *Phys. Rev.* **72,** 914–925 (1947). The Bohr-Wheeler liquid-drop model of nuclear fission has been studied with an electric calculator that allows inclusion of classical deformation energies for more complex deformations. The observed thresholds and spontaneous fission rates are obtained, but no explanation of asymmetric fission is found.

Energy distribution of the fragments resulting from the fission of U^{235} and Th^{232} by slow and by fast neutrons, J. Fowler, L. Rosen (Los Alamos Scientific Laboratory, N. M.), *Phys. Rev.* **72,** 926–930 (1947). The energy spectrum of fission fragments in these reactions was investigated by analyzing the pulses in an ionization chamber with a ten-channel discriminator. The fact there are no extra peaks at the high-energy region indicates that triple fission is less frequent than 1 per thousand.

Natural radiocarbon from cosmic radiation, E. Anderson, *et al.* (Institute for Nuclear Studies and Dept. of Chemistry, Chicago, and Houdry Process Corporation, Marcus Hook, Pa.), *Phys. Rev.* **72,** 931–936 (1947). The amount of C^{14} in biological methane has been measured by concentrating it through thermal diffusion and then introducing it into a brass cylinder counter. The measured activity is consistent with the known neutron flux of cosmic radiation, giving the $N^{14}(n,p)C^{14}$ reaction, and the amount of carbon in exchange equilibrium with the atmosphere.

Ionization of high-energy cosmic-ray electrons, E. Hayward (Univ. of California, Berkeley), *Phys. Rev.* **72,** 937–942 (1947). It is theoretically predicted that the field of an oncoming particle polarizes the medium in such a way as to reduce the ionization, so that above a certain energy the ionization should be practically constant. The apparatus was a cloud chamber with a lateral clearing field in the upper half, to determine the ionization, and lead plates in the lower half, in which the electrons produce showers indicating their energy. The results in most cases are consistent with the theory but sometimes give too high an ionization. This may be due to the coincidence of positron-electron pairs produced by very high-energy gammas.

Extensive air showers and the cascade theory, G. Cocconi (Inst. of Phys., University of Catania, Italy), *Phys. Rev.* **72,** 964–966 (1947). The effects of changing the density of the air (barometric pressure) and of raising the recording apparatus above sea level on the frequency of extensive air showers should be similar, according to cascade theory, since both affect the weight of air traversed. This effect has been experimentally confirmed.

The isotopic constitution of lanthanum and cerium, M. Inghram, R. Haydn, D. Hess (Argonne National Laboratory, Chicago), *Phys. Rev.* **72,** 967–970 (1947). A Nier-type mass spectrometer, 60°, 6-inch radius of curvature, single-focusing, was used. A new isotope of La, 138, of abundance 0.089%, was found. Abundances of Ce isotopes 136, 138, 140, and 142 were 0.193, 0.250, 88.48, and 11.07% respectively. Upper limits for the nonexistence of other isotopes of these elements are given.

On the separation of isotopes in quantity by electromagnetic means, L. Smith, W. Parkins, A. Forrester (Cornell University Ithaca, N. Y.), *Phys. Rev.* **72,** 989–1002 (1947). As inability to produce large numbers of ions is not at present a limitation, the principal problems are separating large quantities of isotopes and retaining sufficient resolution in spite of the large space charge connected with large ion current. By trapping electrons on the axis of the beam in the potential distribution of the ions, a large number can be made to accumulate and effectively reduce the

space charge. In a magnetic field, the electrons can be made even more effective. Velocity modulation or interruption of the beam with electric fields are limited by space charge effects which are discussed. Divergence of charged particles carries lighter ones to greater radial distances, an effect which is suggested as a separation method.

Angular distribution of neutrons from targets bombarded by 190-Mev deuterons, A. Helmholz, E. McMillan, D. Sewell (Univ. of California, Berkeley), (*Phys. Rev.* **72,** 1003–1007 (1947). Small samples of materials subject to neutron activation were placed in contact with the vacuum chamber wall of the 184-inch cyclotron in an array across the beam of neutrons produced by bombardment of target materials from Be to U. The angular distribution is approximately represented by $(1 + a\theta^2)^{-3/2}$ and the half-width in radians by $0.155 + 0.00060Z$, where Z is the atomic number of the target. The neutron intensity was obtained from the activation produced in the samples as measured with a counter.

The production of high-energy neutrons by stripping, R. Serber (Univ. of California, Berkeley), *Phys. Rev.* **72,** 1008–1016 (1947). A theory to explain the production of neutrons by deuteron bombardment of targets is given. The process discussed is one in which the proton of the deuteron collides with the edge of the nucleus and is stripped off, while the neutron misses and continues on. The neutron energy is about half the deuteron energy, with a spread in energy distribution of about 20 Mev. The neutrons emerge in a narrow cone of about 6°, in distinction to the result of direct nuclear encounters, where the cone width is about three times as large. There is an increase in half-width with increasing atomic number (25% from Be to U) because of increased Coulomb field deflection of the deuteron and multiple scattering in the target. If the neutron hits the nucleus, protons are produced by the stripping process. The theory explains the results with the deuteron beam from the 184-inch cyclotron.

Disintegration of the deuteron in flight, S. Dancoff (Univ. of Illinois, Urbana), *Phys. Rev.* **72,** 1017–1022 (1947). This paper considers disintegrations of high-energy deuterons by the Coulomb forces of target nuclei. For 200-Mev deuterons, the cross section is of the order of $2 \times 10^{-29}\ Z^2\ \text{cm}^2$, where Z is the atomic number of the target; neutrons of 75–125 Mev emerge in a beam of 4° half-width. This process gives a smaller cross section than that for direct collision disintegration.

Fluctuations of the number of neutrons in a pile, E. Courant, P. Wallace (Chalk River Laboratory, Ontario, Canada), *Phys. Rev.* **72,** 1038–1048 (1947). Using a probability generating function, calculations are made for the standard deviation of the number of neutrons in a pile which is just below critical and contains distributed neutron sources considering, first, the case of periodic variation of the power level but neglecting delayed neutrons and, second, the case of steady power level but including the effects of delayed neutrons.

Decay scheme of $_{45}Rh^{106}$, W. Peacock (Clinton National Laboratory, Oak Ridge, Tenn.), *Phys. Rev.* **72,** 1049–1053. The beta and gamma rays from Rh^{106} were analyzed, and a scheme devised to explain the results. Beta decay occurs from level 1 to level 4 (energy 3.55 Mev and abundance 82%) and from 1 to 2 (energy 2.30 Mev and abundance 18%). From 2 there is a single gamma ray (energy 1.25 Mev, abundance 1–2%) to 4 and a double gamma emission (energy 0.51 and 0.73 Mev, 17% abundance) to 3

and 4. The two beta spectra seem to be allowed from the values of *ft*, the Fermi function times the partial half-life. Gamow-Teller selection rules seem to be followed, inasmuch as application of Fermi selection rules would require a $0 \rightarrow 0$ gamma ray transition.

Positive excess of slow mesotrons at an altitude of 3.5 km, M. Correll (Ryerson Physical Lab., Univ. of Chicago), *Phys. Rev.* **72,** 1055–1060 (1947). At an altitude of 3.5 km, a cloud chamber was used to investigate the relative numbers of positive and negative mesotrons of momenta between 1.25 and 2.5×10^8 ev/c (selected by an anticoincidence circuit). The mesotrons were distinguished from electrons by multiplication phenomena in lead plates and from protons by their range in the plates. Positive and negative mesotrons were distinguished by a 1670-gauss magnetic field. The results show 47% positive and 53% negative (equal within the experimental error). This differs from the 20% positive excess for mesotrons of energy over 5×10^8 ev. Probably these are formed by a different mechanism.

X-ray emitting isotopes of radioactive Sb and Sn, K. Coleman, M. Pool (Mendenhall Laboratory, Ohio State Univ., Columbus), *Phys. Rev.* **72,** 1070–1074 (1947). Using a bent-crystal spectrometer to investigate the decay and absorption curves of the various activities, the investigators were able to assign 2.8-hour, 5.1-hour, and 39-hour half-lives to Sb-117, 118, and 119 respectively. These isotopes decay by *K*-capture and give off X-rays. Periods of 1.25 and 9 days have been found in Sn.

Energy of beta radiation from S^{35} and C^{14}, A. Solomon, R. Gould, C. Anfinsen (Harvard Medical School, Boston), *Phys. Rev.* **72,** 1097–1100 (1947). A low background thin-window counter was used in comparing the absorption curve in aluminum foils of the S^{35} and C^{14} sources with that of a standard source (RaE). Identical geometry was used. The absorber thicknesses corresponding to various percentages of absorption of the unknown radiation are plotted and the curve extrapolated to find the maximum ranges. The energies computed are 154 ± 4 kev for C^{14} and 167 ± 4 kev for S^{35}.

Nuclear reactions at high energies, R. Serber (Radiation Lab., Univ. of California, Berkeley), *Phys. Rev.* **72,** 1114–1115 (1947). For high-energy incident particles, the nuclear reactions which take place can be described in terms of a "transparent nucleus." That is, the collision can be described in terms of collisions between the incident particle and individual nuclear particles (since the collision time of the high-energy reaction is small compared with the time between collisions among particles in the nucleus). Such a cross section is inversely proportional to the incident energy. The incident particle loses only a small portion of its energy (though in an exchange collision it may change its charge), and transfers momentum perpendicular to its direction of the order of $\hbar/a$ (a is the range of nuclear forces). The momentum transfer is not much larger than the momentum of particles in the nucleus ($\hbar$ divided by the nuclear radius).

In general, what happens depends on the closeness of the incident particle to the center of the struck nucleus. If it grazes, it may make only a single impact and lose only about 25 Mev; if it is a direct impact, a number of collisions may occur and the particle may lose all its energy. Struck particles, having small energy imparted to them, will escape only if they are near the surface. Incident nuclei of more than one particle may be treated as individual particles spatially related.

. . . HAROLD BROWN